

PTO 08-1305

CC=JP DATE=19980106 KIND=A
PN=10003990

ORGANIC ELECTROLUMINESCENT DEVICE
[Yuki EL hakko sochi]

Hiroaki Nakamura, et al.

UNITED STATES PATENT AND TRADEMARK OFFICE
Washington, D.C. December 2007

Translated by: FLS, Inc.

PUBLICATION COUNTRY	(19):	JP
DOCUMENT NUMBER	(11):	10-3990
DOCUMENT KIND	(12):	A
	(13):	
PUBLICATION DATE	(43):	19980106
PUBLICATION DATE	(45):	
APPLICATION NUMBER	(21):	8-174286
APPLICATION DATE	(22):	19960613
ADDITION TO	(61):	
INTERNATIONAL CLASSIFICATION	(51):	H05B 33/14, C09K 11/06
DOMESTIC CLASSIFICATION	(52):	
PRIORITY COUNTRY	(33):	
PRIORITY DATE	(32):	
PRIORITY NUMBER	(31):	
INVENTORS	(72):	HIROAKI NAKAMURA, MASAhide MATSUURA
APPLICANT	(71):	IDEMITSU KOSAN CO LTD
TITLE	(54):	ORGANIC ELECTROLUMINESCENT DEVICE
FOREIGN TITLE	[54A]:	YUKI EL HAKKO SOCHI

[Claims]

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[Claim 1] An organic electroluminescent device, wherein a substrate electrode, organic layers with luminescent layers and an opposing electrode are laminated in successive order on top of a substrate to form a plurality of separate luminescent pixels, wherein the luminescent layers comprise in successive order a blue luminescent layer, a green luminescent layer and a red luminescent layer, wherein

the blue luminescent layer has regions containing a blue fluorescent material in the blue luminescent material, wherein

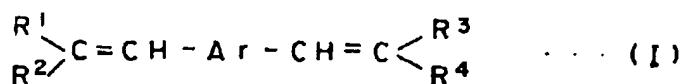
the green luminescent layer has regions containing a green fluorescent material in the blue luminescent material or the green blue luminescent material, and wherein

the red luminescent layer has regions containing a red fluorescent material in the blue luminescent material.

[Claim 2] The organic electroluminescent device of Claim 1, wherein the amount of fluorescent material in the luminescent materials of the various luminescent layers is 0.1 to 10 mol%.

[Claim 3] The organic electroluminescent device of Claim 2, wherein the blue luminescent material is a distylyl arylene compound expressed by Formula (I).

[Formula 1]



* Claim and paragraph numbers correspond to the one in the foreign text.

(In this formula, R^1 through R^4 denote a hydrogen atom, an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted aryl group with 6 to 18 carbon atoms, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted cyclohexyl group, a substituted or unsubstituted aryloxy group with 6 to 18 carbon atoms, and a substituted or unsubstituted pyridyl group. Here, a substituent can be an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, an aryloxy group with 6 to 18 carbon atoms, an acyl group with 1 to 6 carbon atoms, an acyloxy group with 1 to 6 carbon atoms, a carboxyl group, a styryl group, an arylcarbonyl group with 6 to 20 carbon atoms, an aryloxycarbonyl group with 6 to 20 carbon atoms, an alkoxycarbonyl group with 1 to 6 carbon atoms, a vinyl group, an anilino carbonyl group, a carbamoyl group, a phenyl group, a nitro group, a hydroxyl group, or a halogen atom. One or more of these substituents can be used. R^1 through R^4 can be the same or different, and R^1 and R^2 as well as R^3 and R^4 can be bonded with substituents to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. Ar denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms containing one or more substituents, the bonding location being ortho, meta or para. The substituents are the same as above. The substituents of the arylene group can be bonded together to form a substituted or

unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. If Ar is an unsubstituted phenylene, R¹ through R⁴ can denote an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted naphthyl group, a biphenyl group, a cyclohexyl group or an aryloxy group.)

[Claim 4] The organic electroluminescent device in any one of Claims 1 through 3, wherein

either the substrate electrode or the opposing electrode is transparent, and wherein

a color filter layer is formed on the surface of the transparent electrode other than the surface on which the organic layers containing the luminescent layers have been formed.

[Claim 5] The organic electroluminescent device in any one of Claims 1 through 4, wherein in the interface between two of the three regions containing the luminescent material and the fluorescent material, the two opposing regions in the interface include portions contain the same organic compound.

[Claim 6] The organic electroluminescent device in any one of Claims 1 through 5, wherein the thickness of each one of the three regions containing the luminescent material and the fluorescent material is 0.1 to 50 nm.

[Claim 7] The organic electroluminescent device in any one of Claims 1 through 6, wherein the luminescent layers consist of a blue luminescent layer, a green luminescent layer and a red luminescent

layer laminated in this order, and wherein this lamination is repeated more than once.

[Detailed Description of the Invention]

[0001] [Technical Field of the Invention]

The present invention relates to an organic electroluminescent device and, more specifically, to an organic electroluminescent device able to generate white light with high efficiency and high brightness.

[0002] [Prior Art]

Electroluminescent elements (EL elements) are highly visible because of self-luminosity and have superior impact resistance because they are completely solid state. In recent years, various electroluminescent elements have been proposed and implemented using inorganic and organic compounds. Among these elements, the organic electroluminescent elements use much less applied voltage. As a result, various materials and elements are being developed. In recent years, the weight of display elements such as the backlighting and displays themselves has been effectively reduced. The following technologies have been disclosed related to organic electroluminescent elements that generate white light.

(1) Tunnel injection has been used to generate light at the rated energy level of the various layers in an organic electroluminescent laminate structure (European Unexamined Patent Application No. 0,390,551).

(2) A white luminescent element has been developed using the tunnel injection of (1) (JP H3-230584A).

(3) A luminescent layer with a dual-layer structure has been disclosed (JP H2-220390A and JP H2-216790A).

(4) A luminescent layer has been divided into a plurality of sections containing materials with different luminescent wavelengths (JP H4-51491A).

(5) A configuration has been disclosed in which a blue luminescent material (peak fluorescence: 380 nm through 480 nm) and a green luminescent material (480 nm through 580 nm) are laminated, and these contain a red luminescent material (JP H6-207170A).

(6) A configuration has been disclosed in which the blue luminescent layer contains a blue fluorescent material, the green luminescent layer has a region containing a red fluorescent material, and both contain a green fluorescent material (JP H7-142169A).

[0003] [Problems Solved by the Invention]

These technologies experience the following problems.

(1) In European Unexamined Patent Application No. 0,390,551, tunnel injection occurs due to the accumulation of a carrier in the carrier interface. As a result, regional value voltage is used to generate white light. Below this regional value, white light is not generated. This makes gradation impossible.

(2) Because of the mixed luminescence from fluorescent materials with two colors in JP H3-230584A, the white color is poor.

(3) White luminescence occurs in JP H2-216790A, but the brightness is 110 cd/m² at an applied voltage of 30 V. In other words, the luminance efficiency is low for such a high operating voltage.

(4) In JP H4-51491A, the structure is luminescent only on the end surface and so cannot be used in a panel.

(5) There are two luminescent layers in JP H6-207170A and both have low luminescent efficiency. The purity of the white light is also poor.

(6) There are two luminescent layers in JP H7-142169A and both have low luminescent efficiency.

[0004] In light of these problems, the purpose of the present invention is to provide an organic electroluminescent device able to efficiently generate white light with high brightness.

[0005] [Means of Solving the Problems]

In order to achieve this purpose, the present invention is an organic electroluminescent device, wherein

a substrate electrode, organic layers with luminescent layers and an opposing electrode are laminated in successive order on top of a substrate to form a plurality of separate luminescent pixels, wherein

the luminescent layers comprise in successive order a blue luminescent layer, a green luminescent layer and a red luminescent layer, wherein

the blue luminescent layer has regions containing a blue fluorescent material in the blue luminescent material, wherein

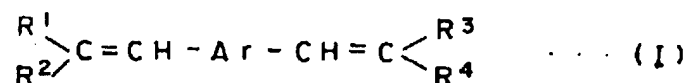
the green luminescent layer has regions containing a green fluorescent material in the blue luminescent material or the green blue luminescent material, and wherein

the red luminescent layer has regions containing a red fluorescent material in the blue luminescent material.

[0006] In a preferred embodiment, the amount of fluorescent material in the luminescent materials of the various luminescent layers is 0.1 to 10 mol%.

[0007] Also, in a preferred embodiment, the blue luminescent material in the organic electroluminescent device is a distyryl arylene compound expressed by Formula (I).

[0008] [Formula 1]



[0009] (In this formula, R^1 through R^4 denote a hydrogen atom, an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted aryl group with 6 to 18 carbon atoms, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted cyclohexyl group, a substituted or unsubstituted aryloxy group with 6 to 18 carbon atoms, and a substituted or unsubstituted pyridyl group. Here, a substituent can be an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, an aryloxy group with 6 to 18 carbon atoms, an acyl group with 1 to 6 carbon atoms, an acyloxy group with 1 to 6 carbon atoms, a carboxyl group, a styryl group, an arylcarbonyl group with 6 to 20 carbon atoms, an aryloxy carbonyl group with 6 to 20 carbon atoms, an alkoxycarbonyl group with 1 to 6 carbon atoms, a vinyl group, an anilinocarbonyl

group, a carbamoyl group, a phenyl group, a nitro group, a hydroxyl group, or a halogen atom. One or more of these substituents can be used. R^1 through R^4 can be the same or different, and R^1 and R^2 as well as R^3 and R^4 can be bonded with substituents to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. Ar denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms containing one or more substituents, the bonding location being ortho, meta or para. The substituents are the same as above. The substituents of the arylene group can be bonded together to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. If Ar is an unsubstituted phenylene, R^1 through R^4 can denote an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted naphthyl group, a biphenyl group, a cyclohexyl group or an aryloxy group.)

[0010] Also, in a preferred embodiment, either the substrate electrode or the opposing electrode is transparent, and a color filter layer is formed on the surface of the transparent electrode other than the surface on which the organic layers containing the luminescent layers have been formed. Also, in a preferred embodiment, in the interface between two of the three regions containing the luminescent material and the fluorescent material, the two opposing regions in the interface include portions contain the same organic compound. Also, in a preferred embodiment, the thickness of each one of the three regions

containing the luminescent material and the fluorescent material is 0.1 to 50 nm.

[0011] Also, in a preferred embodiment, the luminescent layers consist of a blue luminescent layer, a green luminescent layer and a red luminescent layer laminated in this order, and wherein this lamination is repeated more than once.

[0012] [Embodiment of the Invention]

The following is a detailed explanation with reference to the drawing of an embodiment of the organic electroluminescent device of the present invention. FIG 1 is a simplified cross-sectional view of an embodiment of the present invention. As shown in FIG 1, the organic electroluminescent device of the present invention consists of a substrate electrode 20, organic layers 30 including the luminescent layers and an opposing electrode 40 laminated in successive order on top of a substrate 10 to form a plurality of separate luminescent pixels. Here, a luminescent pixel consists of a substrate electrode 20, an opposing electrode 40 and organic layers 30 including luminescent layers interposed between them. If necessary, a color filter can be installed to realize the desired color. Here, the pixels can be controlled to light and extinguish them individually. The luminescent pixels in FIG 1 are arranged on an X-Y matrix, and the desired pixels can be lit to display a pattern or character.

[0013] The luminescent layers comprise in successive order a blue luminescent layer 31, a green luminescent layer 32 and a red luminescent layer 33. The blue luminescent layer 31 has regions

containing a blue fluorescent material in the blue luminescent material, the green luminescent layer 32 has regions containing a green fluorescent material in the blue luminescent material or the green blue luminescent material, and the red luminescent layer 33 has regions containing a red fluorescent material in the blue luminescent material.

[0014] Here, a blue luminescent material is an organic compound with a solid-state fluorescent peak wavelength between 380 nm and 480 nm, a green luminescent material is an organic compound with a solid-state fluorescent peak wavelength between 480 nm and 580 nm, a blue fluorescent material is an organic compound with a solution-state fluorescent peak wavelength between 380 nm and 480 nm, a green fluorescent material is an organic compound with a solution-state fluorescent peak wavelength between 480 nm and 580 nm, and a red luminescent material is an organic compound with a solution-state fluorescent peak wavelength between 580 nm and 650 nm.

[0015] Also, the amount of fluorescent material in the luminescent materials of the various luminescent layers is 0.1 to 10 mol%, preferably 0.3 to 5 mol%. If less than 0.1 mol%, the luminescence intensity of the fluorescent material is insufficient. If greater than 10 mol%, concentration quenching of the fluorescence occurs, the luminescent intensity decreases and the luminescence wavelength changes.

[0016] The thickness of the three regions containing the luminescent material and the fluorescent material is 0.1 to 50 nm, and

preferably 5 to 40 nm. If less than 0.1 nm, a thin film cannot be formed. If greater than 50 nm, the operating voltage increases.

[0017] In the interface between two of the three regions containing the luminescent material and the fluorescent material, the two opposing regions in the interface include portions should contain the same organic compound. In other words, when the green luminescent layer consists of a green fluorescent material in the blue luminescent material, the same material is selected as the luminescent material to reduce the time and effort required to manufacture the element.

[0018] In addition to the luminescent layers 31, 32 and 33 serving as the organic layers 30, a hole-injecting layer 34 and an electron-injecting layer 35 are arranged in the organic electroluminescent element of this embodiment. Layers 34 and 35 can consist of two or more layers. In this embodiment, the luminescent layers 31, 32, 33 are a B/G/R laminate. However, this can be repeated more than once to obtain a thicker laminate (e.g., B/G/R/B/G/R, etc.). The B/G/R laminate order is B/G/R from the light discharge side. This is preferred from the standpoint of less light reabsorption.

[0019] The following is a detailed explanation of the configurational elements used in an organic electroluminescent device of the present invention.

[0020] 1. Substrate

The substrate used in the present invention should be made from a material that is transparent and sufficiently rigid to support a multicolor luminescent device. If the gap between the organic

electroluminescent device and the fluorescent layer is too great in a precision display, the light emitted from the organic luminescent device is absorbed by the fluorescent layer and the desired color is not obtained. The viewing angle is also reduced. For this reason, the thickness of the transparent insulating layer has to be reduced. If the thickness is reduced, the mechanical strength (e.g., the impact resistance) of the multicolor luminescent device is weakened. In the present invention, the substrate reinforces the multicolor luminescent device and improves the mechanical strength (e.g., the impact resistance).

[0021] Specific examples of materials include a glass plate, ceramic plate, plastic plate (polycarbonate, acrylic, vinyl chloride, polyethylene terephthalate, polyimide, polyester, etc.), metal plate, or a plate consisting of the same material as the insulating layer described below. Metal is preferred because of its superior mechanical strength such as impact resistance. The use of metal allows the thickness of the transparent insulating film to be reduced. Even when a metal substrate is thin, it still provides an adequate reinforcing effect. Preferred examples include any metal or an alloy of any metal selected from among stainless steel, iron, copper, aluminum, magnesium, nickel, zinc, chromium, titanium, molybdenum, silicon, germanium or tantalum. The material should have superior processing properties during etching for void formation, should be sufficiently rigid as a substrate, and should be an inexpensive material from the standpoint of cost. When voids are formed, the luster of the metal can appear

through the side surfaces of the voids. This reflects or scatters the light emitted from the fluorescent layers, and allows a multicolor luminescent device to be obtained with efficient luminescence and superior visibility. There are no limitations on the thickness of the substrate. However, the voids in a precision multicolor luminescent display have to be formed with precision. Also, the substrate has to be rigid enough to support a multicolor luminescent display while being as thin as possible. This normally ranges between 5 μm and 5 mm, preferably between 7 μm and 700 μm , and ideally between 10 μm and 300 μm .

[0022] 2. Substrate Electrode

The substrate electrode should be made from a metal, alloy, conductive compound or mixture with a high work function ($> 4 \text{ eV}$). Specific examples of electrode materials include metals such as Au, and conductive transparent materials such as CuI, ITO, SnO_2 and ZnO . The substrate electrode can be formed by creating a thin film from one of these electrode materials using a method such as vapor deposition or sputtering. If the luminescence of the luminescent layers is to be emitted from the substrate, the transmissivity of the substrate electrode to light should be greater than 10%. The sheet resistance of the substrate electrode should be several hundred Ω/\square [sic]. The thickness of the substrate electrode depends on the material. It usually ranges between 10 nm and 1 μm , and preferably between 10 and 200 nm.

[0023] 3. Opposing Electrode

The opposing electrode should be made from a metal, alloy, conductive compound or mixture with a low work function (< 4 eV). Specific examples of electrode materials include sodium, sodium-potassium alloys, magnesium, lithium, magnesium silver alloys, aluminum and aluminum oxide (Al_2O_3), aluminum-lithium alloys, indium and rare earth metals. The opposing electrode can be formed by creating a thin film from one of these electrode materials using a method such as vapor deposition or sputtering. The sheet resistance of the opposing electrode should be several hundred Ω/\square [sic]. The thickness of the opposing electrode depends on the material. It usually ranges between 10 nm and 1 μm , and preferably between 50 and 200 nm. In an organic electroluminescent element of the present invention, either the substrate electrode or the opposing electrode is transparent or semi-transparent. They should be transmissive to the generated light in order to increase the luminescent efficiency.

[0024] In the present invention, either the substrate electrode or the opposing electrode is transparent to light in order to let the generated light pass through. A color filter or black matrix can be installed on the side of the transparent electrode other than the side on which the organic layers containing luminescent layers are formed in order to improve the color purity.

[0025] The color filter used in the present invention can be one of the following pigments. These pigments can be dissolved or dispersed in a binder resin in solid state.

[0026] Red (R) Pigments: Perylene pigments, lake pigments, azo pigments, quinacridone pigments, anthraquinone pigments, anthracene pigments, isoindoline pigments, isoindolinone pigments. These can be used alone or in mixtures of two or more.

[0027] Green (G) Pigments: Halogen multi-substituted phthalocyanine pigments, halogen multi-substituted copper phthalocyanine pigments, triphenylmethane basic dyes, isoindoline pigments, isoindolinone pigments. These can be used alone or in mixtures of two or more.

[0028] Blue (B) Pigments: Copper phthalocyanine pigments, indanthrone pigments, indophenol pigments, cyanine pigments, dioxazine pigments. These can be used alone or in mixtures of two or more.

[0029] The binder resin should be a transparent (>50% visible light) material. Examples of transparent resins (polymers) include polymethylmethacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinylpyrrolidone, hydroxyethyl cellulose, and carboxymethyl cellulose.

[0030] Because a color filter is arranged separately in planar fashion, a photosensitive resin appropriate for photolithography should also be selected. Examples of photocured resist materials with reactive vinyl groups include acrylic acid, methacrylic acid, polyvinylcinnamate, and cyclic rubber materials. If a printing method is used, a printing ink (medium) using a transparent resin should be selected. Examples of transparent resins include monomers, oligomers and polymers of polyvinyl chloride resins, melamine resin, phenol

resin, alkyd resin, epoxy resin, polyurethane resins, polyester resins, maleic acid resins, and polyamide resin; as well as polymethyl methacrylates, polyacrylates, polycarbonates, polyvinyl alcohol, polyvinylpyrrolidone, hydroxyethyl cellulose, and carboxymethyl cellulose.

[0031] If the color filter consists mainly of a pigment, a thin film can be formed using vacuum deposition or sputtering via a mask with the desired color filter pattern. If the color filter consists of a pigment and a binder resin, a mixture of a fluorescent pigment, resin and resist can be dispersed or dissolved and a thin film formed using a method such as spin coating, roller coating or casting. The photolithography method can also be used to create a color filter with the desired pattern, or a printing method can be used to create a color filter with the desired pattern.

[0032] The following is the preferred thickness and transmissivity for the various color filters.

R: Film Thickness 0.5-5.0 μm (Transmissivity >50%/610 nm), G: Film Thickness 0.5-5.0 μm (Transmissivity >50%/545 nm), B: Film Thickness 0.2-5.0 μm (Transmissivity >50%/460 nm)

[0033] In a color filter consisting of a pigment and a binder resin, the pigment density can be in any range that does not cause a problem with the color filter, that allows for patterning, and that allows the light generated by the organic electroluminescent element to pass through sufficiently. This depends on the pigment type, but 5-

50 wt% color pigment can be included in a color filter film including a binder resin.

[0034] The black matrix used in the present invention can be a metal or metal oxide thin film or a black pigment. Specific examples of metal and metal oxide thin films include thin films of metals such as chromium (Cr), nickel (Ni) and copper (Cu) as well as their oxides. A mixture of a metal and metal oxide with an optical density greater than 3.0 (film thickness: 100-300 Å) is preferred.

[0035] Specific examples of black pigments include carbon black, titanium black, aniline black or a mixture of color filter pigments. A solid-state black color can be obtained by dissolving or dispersing the pigments in a binder resin as in the case of a color filter.

[0036] A metal or metal oxide thin film can be formed over the entire surface of an insulating substrate using the sputtering method, deposition method or CVD method, and over the entire surface of the display unit by forming a thin film using a masking technique and then patterning the film using photolithography to form a plastic matrix pattern.

[0037] If a black pigment is used, it can be patterned in the same manner as a color filter to obtain a black matrix.

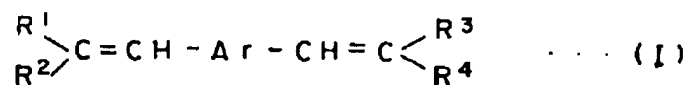
[0038] 4. Organic Layer Containing Luminescent Layers

(1) Luminescent Layers

There are no restrictions on the organic compound used in the blue luminescent material. Examples include the organic compounds in JP H3-231970A, WO92/05131A, Japanese Patent Application No. 5-170354

and Japanese Patent Application No. 5-129438 that satisfy the fluorescent conditions of a blue luminescent material. Preferred examples include the organic compounds in JP H3-231970A, WO92/05131A and Japanese Patent Application No. 5-170345 that satisfy the fluorescent conditions of a blue luminescent material, combinations with some of the appropriate compounds disclosed in Japanese Patent Application No. 5-129438, and compounds used in the hole-introducing transfer layer. All of these must satisfy the fluorescent conditions of a blue luminescent material. Examples of compound that satisfy the fluorescent conditions of a blue luminescent material described in JP3-231970A and WO92/05131A include the distylyl arylene compounds expressed by Formula (I)

[0039] [Formula 1]



[0040] (In this formula, R^1 through R^4 denote a hydrogen atom, an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted aryl group with 6 to 18 carbon atoms, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted cyclohexyl group, a substituted or unsubstituted aryloxy group with 6 to 18 carbon atoms, and a substituted or unsubstituted pyridyl group. Here, a substituent can be an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, an aryloxy group with 6 to 18 carbon atoms, an acyl group with 1 to 6 carbon

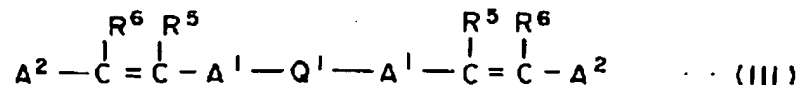
atoms, an acyloxy group with 1 to 6 carbon atoms, a carboxyl group, a stylyl group, an arylcarbonyl group with 6 to 20 carbon atoms, an aryloxy carbonyl group with 6 to 20 carbon atoms, an alkoxycarbonyl group with 1 to 6 carbon atoms, a vinyl group, an anilinocarbonyl group, a carbamoyl group, a phenyl group, a nitro group, a hydroxyl group, or a halogen atom. One or more of these substituents can be used. R^1 through R^4 can be the same or different, and R^1 and R^2 as well as R^3 and R^4 can be bonded with substituents to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. Ar denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms containing one or more substituents, the bonding location being ortho, meta or para. The substituents are the same as above. The substituents of the arylene group can be bonded together to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring. If Ar is an unsubstituted phenylene, R^1 through R^4 can denote an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted naphthyl group, a biphenyl group, a cyclohexyl group or an aryloxy group.), the aromatic methilidine compounds expressed by Formula (II)

A-Q-B ... (II)

(In this formula, A and B are the same or different and denote a univalent group in which a single hydrogen atom has been removed from a compound expressed by Formula (I). Here, Q denotes a conjugate

severed at the divalent bond.), or an aromatic dimethidine compound expressed by Formula (III)

[0041] [Formula 2]



[0042] (In this formula, A^1 denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms or a divalent aromatic heterocyclic group. The bonding position can be ortho, meta or para. A^2 denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms or a univalent aromatic heterocyclic group. R^5 and R^6 denote a hydrogen atom, a substituted or unsubstituted arylene group with 6 to 20 carbon atoms, a cyclohexyl group, a univalent aromatic heterocyclic group, an alkyl group with 1 to 10 carbon atoms, an aralkyl group with 7 to 20 carbon atoms, or an alkoxy group with 1 to 10 carbon atoms. R^5 and R^6 can be the same or different. Here, the substituent can be an alkyl group, an aryloxy group, an amino group or a phenyl group with or without any of these groups. There can be one or more substituents. The substituents for R^5 can be bonded with A^1 to form a saturated or unsaturated five-member or six-member ring. Similarly, The substituents for R^6 can be bonded with A^2 to form a saturated or unsaturated five-member or six-member ring. Also, Q^1 is the same as described above.)

[0043] Here, R^1 through R^4 in Formula (I) are the same or different, and denote a hydrogen atom, an alkyl group with 1 to 6 carbon atoms (a methyl group, ethyl group, n-propyl group, isopropyl group, n-butyl group, isobutyl group, sec-butyl group, tert-butyl

group, isopentyl group, t-pentyl group neopentyl group, isoheptyl group, etc.), an alkoxy group with 1 to 6 carbon atoms (a methoxy group, ethoxy group, propoxy group, butoxy group, etc.), an aralkyl group with 1 to 8 carbon atoms (a benzyl group, phenethyl group, etc.), an aryl group with 6 to 18 carbon atoms (a phenyl group, biphenyl group, naphthyl group, etc.), a cyclohexyl group, an aromatic heterocyclic group (pyridyl group, quinolyl group), or an aryloxy group with 6 to 8 carbon atoms (a phenoxy group, biphenyloxy group, naphthyloxy group, etc.).

[0044] R^1 through R^4 can also be bonded to a substituent. In other words, R^1 through R^4 can denote a phenyl group containing a substituent, an aralkyl group containing a substituent, a cyclohexyl group containing a substituent, a biphenyl group containing a substituent, or a naphthyl group containing a substituent. Here, the substituent can be an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, an aryloxy group with 6 to 18 carbon atoms, an acyl group with 1 to 6 carbon atoms, an acyloxy group with 1 to 6 carbon atoms, a carboxyl group, a styryl group, an arylcarbonyl group with 6 to 20 carbon atoms, an aryloxycarbonyl group with 6 to 20 carbon atoms, an alkoxycarbonyl group with 1 to 6 carbon atoms, a vinyl group, an anilinocarbonyl group, a carbamoyl group, a phenyl group, a nitro group, a hydroxyl group or a halogen atom. One or more substituents can be used. Examples of aralkyl groups containing a substituent include alkyl-substituted aralkyl groups (a methyl benzyl group,

methylphenethyl group, etc.), alkoxy-substituted aralkyl groups (a methoxybenzyl group, ethoxyphenethyl group, etc.), aryloxy-substituted aralkyl groups (a phenoxybenzyl group, naphthyloxyphenethyl group, etc.), and phenyl-substituted aralkyl groups (a phenylphenethyl group etc.). Examples of phenyl group containing a substituent include an alkyl-substituted phenyl group (a tolyl group, dimethylphenyl group, ethylphenyl group, etc.), an alkoxy-substituted phenyl group (a methoxyphenyl group, ethoxyphenyl group, etc.), an aryloxy-substituted phenyl group (a phenoxyphenyl group, naphthyloxyphenyl group, etc.), and a phenyl-substituted phenyl group (i.e., a biphenyl group). Examples of cyclohexyl groups containing a substituent include an alkyl-substituted cyclohexyl group (a methylcyclohexyl group, dimethylcyclohexyl group, ethylcyclohexyl group, etc.), an alkoxy-substituted cyclohexyl group (a methoxycyclohexyl group, ethoxycyclohexyl group, etc.), an aryloxy-substituted cyclohexyl group (a phoxycyclohexyl group, naphthyloxycyclohexyl group, etc.), and a phenyl-substituted cyclohexyl group (a phenylcyclohexyl group). Examples of naphthyl groups containing substituents include an alkyl-substituted naphthyl group (a methylnaphthyl group, dimethylnaphthyl group, etc.), an alkoxy-substituted naphthyl group (a methoxynaphthyl group, ethoxynaphthyl group, etc.), an aryloxy-substituted naphthyl group (a phenoxynaphthyl group, naphthyloxynaphthyl group), a phenyl-substituted naphthyl group (phenylnaphthyl group).

[0045] Preferred examples for R^1 through R^4 include an alkyl group with 1 to 6 carbon atoms, an aryloxy group, a phenyl group, a

naphthyl group, a biphenyl group or a cyclohexyl group. These can be substituted or unsubstituted. R^1 through R^4 can be the same or different, and R^1 and R^2 as well as R^3 and R^4 can be bonded with substituents to form a substituted or unsubstituted saturated or unsaturated five-member ring, or a substituted or unsubstituted saturated or unsaturated six-member ring.

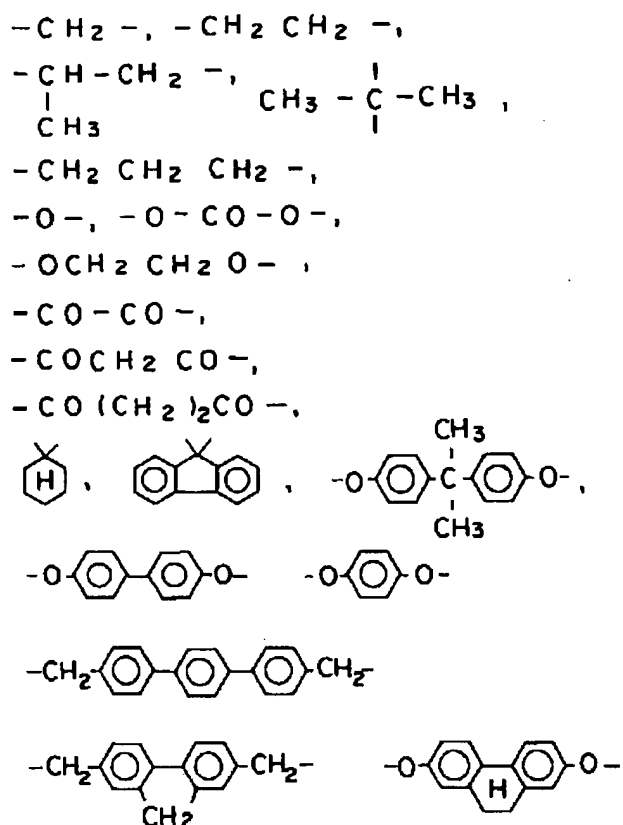
[0046] In Formula (I), Ar denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms; a substituted or unsubstituted arylene group such as a phenylene group, biphenylene group, p-terphenylene group, naphthalene group, terphenylene group, naphthalenediyl group, anthracenediyl group, phenanthrenediyl group or phenalenediyl group. The methyldine ($=C=CH-$) bonding location can be ortho, meta or para. If Ar is an unsubstituted phenylene, R^1 through R^4 denote an alkoxy group with 1 to 6 carbon atoms, an aralkyl group with 7 to 18 carbon atoms, a substituted or unsubstituted naphthyl group, a biphenyl group, a cyclohexyl group or an aryloxy group. The substituent can be an alkyl group (methyl group, ethyl group, n-propyl group, isopropyl group, n-butyl group, isobutyl group, sec-butyl group, t-butyl group, isopentyl group, neopentyl group, or isohexyl group), an alkoxy group (methoxy group, ethoxy group, propoxy group, isopropoxy group, butyloxy group, isobutyloxy group, sec-butyloxy group, t-butyloxy group, isopentyloxy group, t-pentyloxy group), an aryloxy group (phenoxy group, naphthyloxy group), an acyl group (formyl group, acetyl group, propionyl group, butyryl group), acyloxy group, aralkyl group (benzyl group, phenethyl group), phenyl group,

hydroxyl group, carboxyl group, anilinocarbonyl group, carbamoyl group, aryloxy carbonyl group, methoxycarbonyl group, ethoxycarbonyl group, butoxycarbonyl group, nitro group, or a halogen atom. One or more substituents can be used.

[0047] The methyldine aromatic compound expressed by Formula (I) has two methyldine groups ($=CH=CH-$) in a single molecule, and there are four combinations based on the geometric isomerism of the methyldine groups: cis-cis, trans-cis, cis-trans, and trans-trans. The blue fluorescent material in the electroluminescent element of the present invention is a mixture of any of these and a mixture of any geometric isomerisms. However, all trans is preferred. The substituents can be bonded to each other to form a substituted or unsubstituted saturated or unsaturated five-member ring or six-member ring.

[0048] A and B in Formula (II) are the same or different and denote a univalent group in which a single hydrogen atom has been removed from a compound expressed by Formula (I). Here, Q in Formula (II) denotes a conjugate severed at the divalent bond. Here, the conjugate includes the non-polarity of the π -electron, a conjugate double bond, and an unpaired electron or orphan electron pair. The following are specific examples of Q.

[0049] [Formula 3]



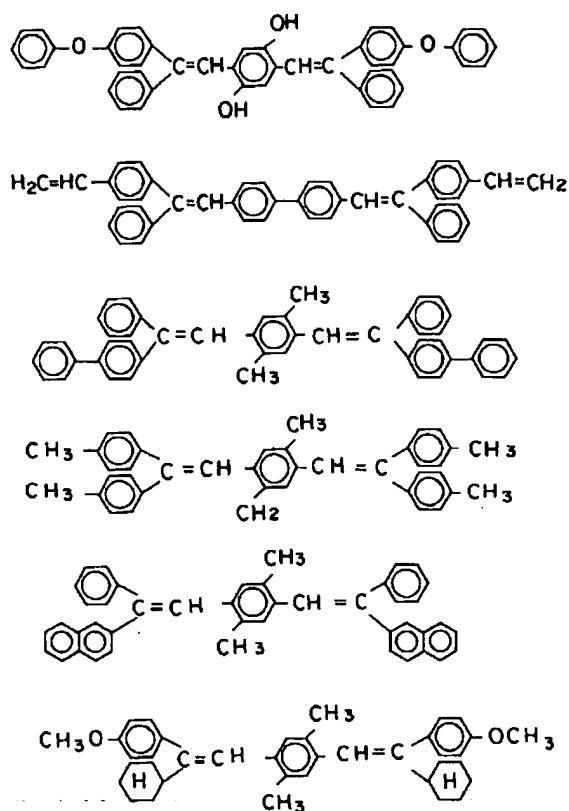
[0050] A conjugate severed at the divalent bond is used because A and B [i.e., compounds in Formula (I)] are used alone for the electroluminescent color obtained from an organic electroluminescent element of the present invention, and a compound expressed by Formula (II) is used so that the electroluminescent color obtained from an organic electroluminescent element of the present invention does not change. In other words, this keeps the wavelength of the blue fluorescence expressed by Formula (I) and Formula (II) from becoming longer or shorter. When the conjugate is continuously severed at the divalent group, the glass transition temperature (T_g) rises, a uniform, pinhole-free fine crystalline or amorphous thin film can be obtained,

and the uniformity of the luminescence is improved. By bonding the conjugate severed at the divalent group, the wavelength of the electroluminescence does not become longer, and synthesis and refining are easier.

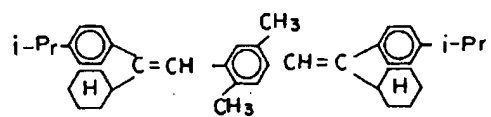
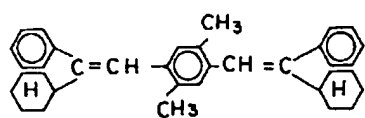
[0051] In Formula (III), A^1 denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms or a divalent aromatic heterocyclic group, and A^2 denotes a substituted or unsubstituted arylene group with 6 to 20 carbon atoms (phenyl group, biphenyl group, naphthyl group, etc.) or a univalent aromatic heterocyclic group. R^5 and R^6 denote a hydrogen atom, a substituted or unsubstituted arylene group with 6 to 20 carbon atoms, a cyclohexyl group, a univalent aromatic heterocyclic group, an alkyl group with 1 to 10 carbon atoms (methyl group, ethyl group, n-propyl group, isopropyl group, n-butyl group, isopropyl group, sec-butyl group, tert-butyl group, isopentyl group, t-pentyl group, neopentyl group, isopentyl group, etc.), an aralkyl group with 7 to 20 carbon atoms (benzyl group, phenethyl group, etc.), or an alkoxy group with 1 to 10 carbon atoms (methoxy group, ethoxy group, propoxy group, butoxy group, etc.). R^5 and R^6 can be the same or different. Here, the substituent can be an alkyl group, an aryloxy group, an amino group or a phenyl group with or without any of these groups. There can be one or more substituents. The substituents for R^5 can be bonded with A^1 to form a saturated or unsaturated five-member or six-member ring. Similarly, The substituents for R^6 can be bonded with A^2 to form a saturated or unsaturated five-member or six-member ring. Also, Q is a conjugate

severed at the divalent group as described above. In addition, the A¹ bond can be an ortho, meta or para bond. In the present invention, organic compounds expressed by Formula (I), Formula (II) or Formula (III) have to be compounds that manifest blue-violet, violet-blue, blue, green-blue or blue-green in the CIE chromacity coordinates. Specific examples include the following.

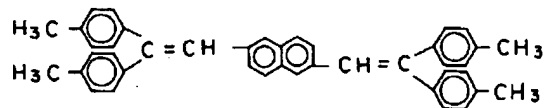
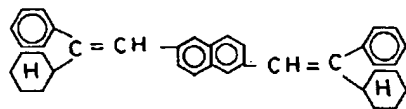
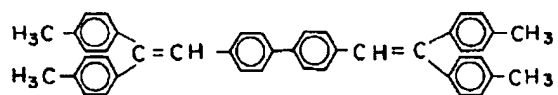
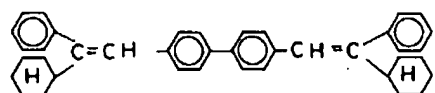
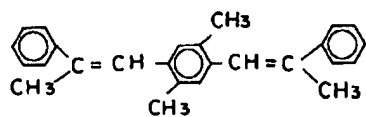
[0052] [Formula 4]



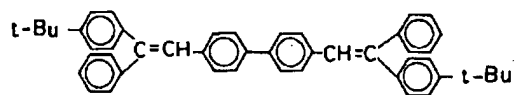
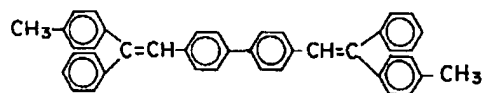
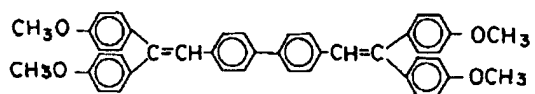
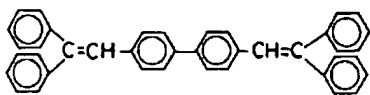
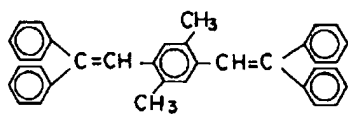
[0053] [Formula 5]



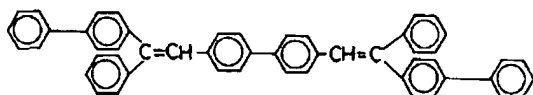
i-Pr : Isopropyl (Same Below)



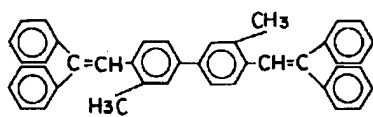
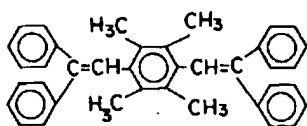
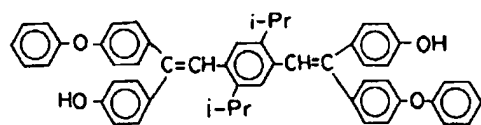
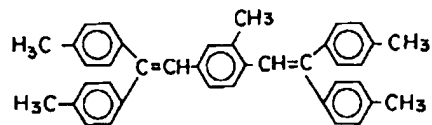
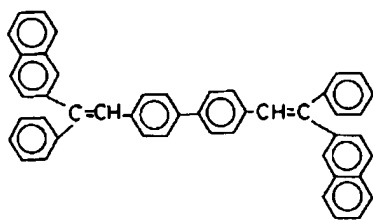
[0054] [Formula 6]



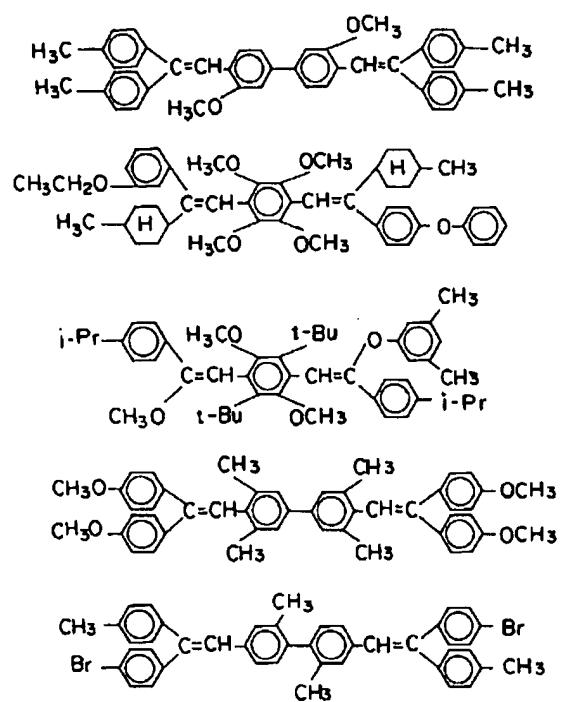
t-Bu: t-Butyl (Same Below)



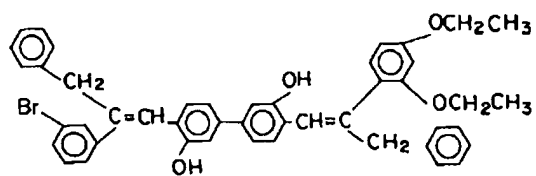
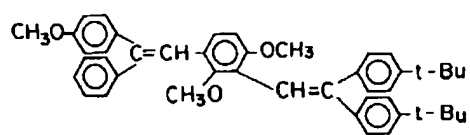
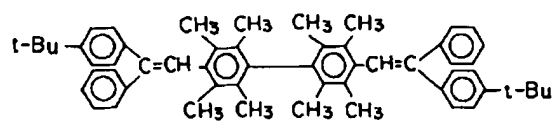
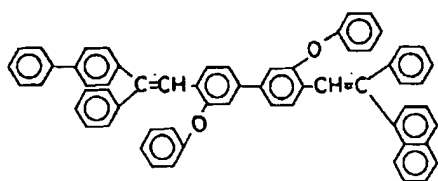
[0055] [Formula 7]



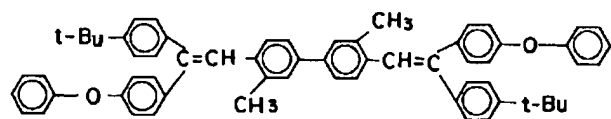
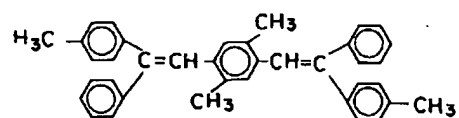
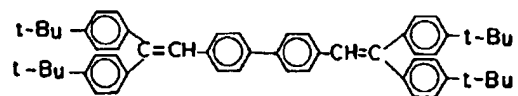
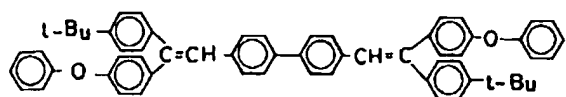
[0056] [Formula 8]



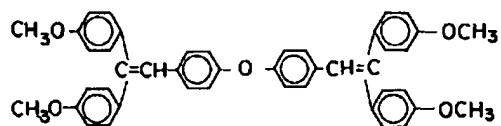
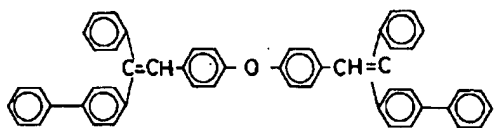
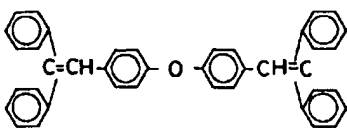
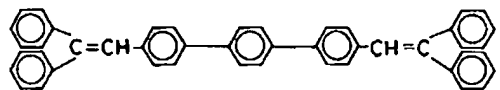
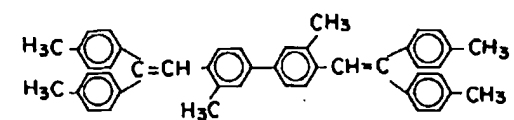
[0057] [Formula 9]



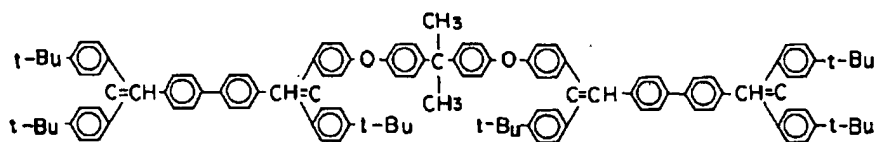
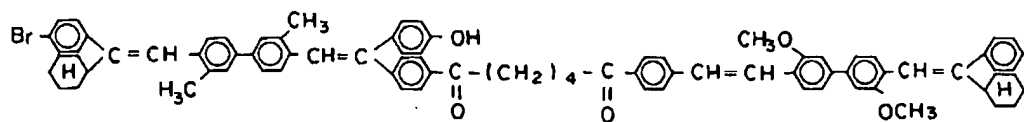
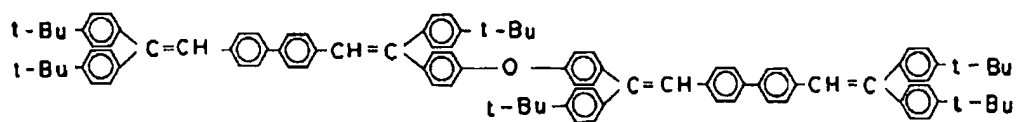
[0058] [Formula 10]



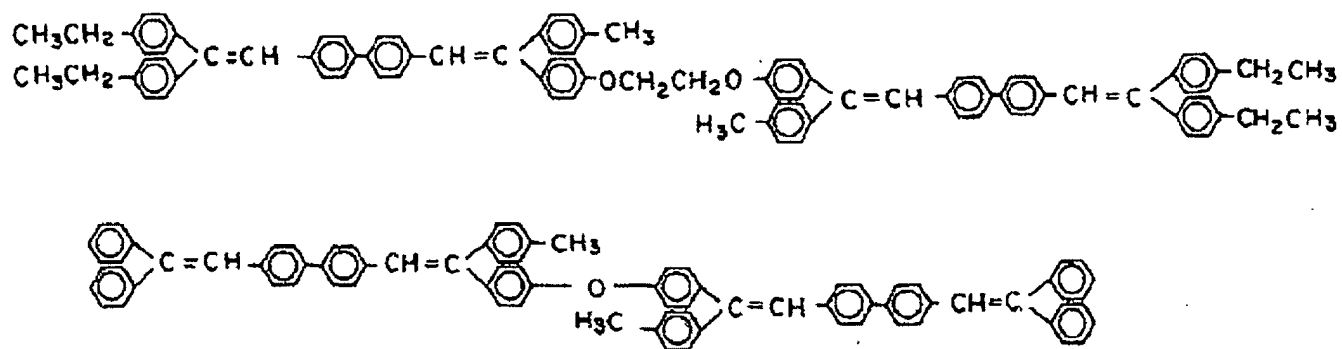
[0059] [Formula 11]



[0060] [Formula 12]

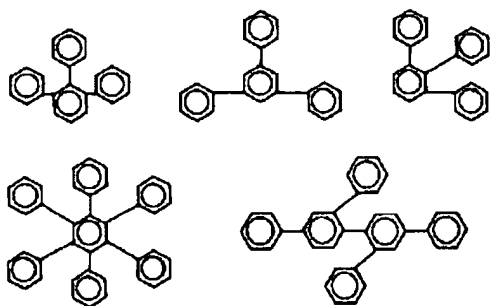


[0061] [Formula 13]



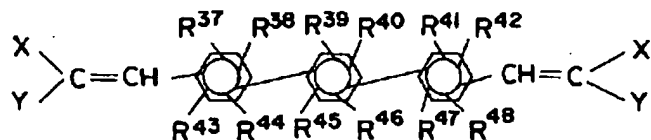
[0062] Other organic compounds include the following.

[0063] [Formula 14]



[0064] A compound satisfying the fluorescence conditions for a blue-green fluorescent material described in the specification of Japanese Patent Application No. 5-170354 is a styryl compound of a terphenylene derivative expressed by Formula (XI).

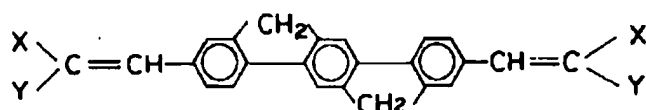
[0065] [Formula 15]



[0066] (In this formula, R^{37} through R^{48} separately denote a hydrogen atom or an alkyl group with 1 to 6 carbon atoms. At least one of R^{37} through R^{48} is an alkyl group with 1 to six carbon atoms. R^{38} and R^{39} , R^{40} and R^{41} , R^{44} and R^{45} , and R^{46} and R^{47} can be bonded together to form a saturated or unsaturated five-member or six-member ring. X and Y separately denote substituted or unsubstituted aryl groups with 6 to 20 carbon atoms. X and Y can be bonded together to form a saturated or unsaturated five-member or six-member ring. Here, a substituent can be an alkyl group with 1 to 6 carbon atoms, an alkoxy group with 1 to 6 carbon atoms, an aryloxy group with 6 to 18 carbon atoms, a phenyl group, an amino group, a cyano group, a nitro group, a hydroxyl group or a halogen atom. There can be one or more substituents.)

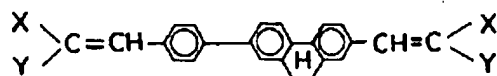
[0067] In Formula (XI), R^{37} through R^{48} separately denote a hydrogen atom or an alkyl group with 1 to 6 carbon atoms such as a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a sec-butyl group, a t-butyl group, an isopentyl group, a t-pentyl group, a neopentyl group, an n-hexyl group, or an isohexyl group. At least one of R^{37} through R^{48} is an alkyl group with 1 to six carbon atoms, preferably a methyl group or an ethyl group. R^{38} and R^{39} , R^{40} and R^{41} , R^{44} and R^{45} , and R^{46} and R^{47} can be bonded together to form a saturated or unsaturated five-member or six-member ring. Among the examples of styryl compounds with a saturated or unsaturated five-member or six-member ring, the following formula expresses a specific example in which R^{38} and R^{39} as well as R^{40} and R^{41} form a saturated five-member ring.

[0068] [Formula 16]



[0069] The following formula expresses a specific example in which R⁴⁶ and R⁴⁷ form a saturated five-member ring.

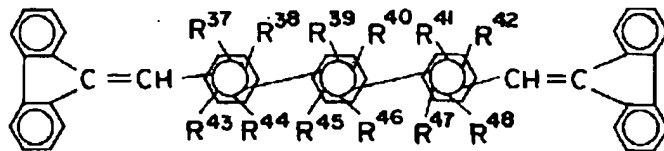
[0070] [Formula 17]



[0071] X and Y separately denote a substituted or unsubstituted aryl group with 6 to 20 carbon atoms such as a phenyl group, naphthyl group, biphenyl group, terphenyl group, anthralyl group, phenanthryl group, pyrenyl group or perilenyl group. Here, a substituent can be an alkyl group with 1 to 6 carbon atoms such as a methyl group, ethyl group, n-propyl group, isopropyl group, n-butyl group, isobutyl group, sec-butyl group, t-butyl group, isopentyl group, t-pentyl group, neopentyl group or isopentyl group; an alkoxy group with 1 to 6 carbon atoms such as a methoxy group, ethoxy group, propoxy group, n-propoxy group, isopropoxy group, n-butyloxy group, isobutyloxy group, sec-butyloxy group, isopentyloxy group, t-pentyloxy group or n-hexyloxy group; a phenyl group; an amino group; a cyano group; a nitro group; a hydroxyl group; or a halogen atom. There can be one or more substituents. X and Y can be bonded together to form a saturated or unsaturated five-member or six-member ring. Among styryl groups with a saturated or unsaturated five-member or six-member group, compounds in

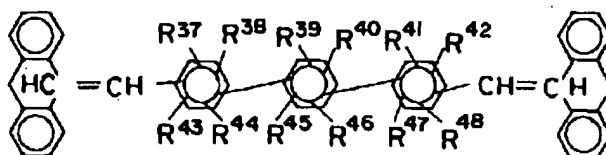
which X and Y form a saturated five-member group are expressed by the following formula.

[0072] [Formula 18]



[0073] Compounds in which X and Y form a saturated six-member group are expressed by the following formula.

[0074] [Formula 19]

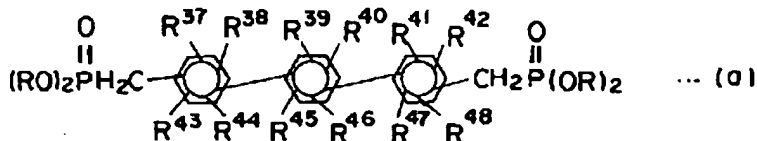


[0075]

[0076] A styryl compound expressed by Formula XI can be manufactured using any method common in the art. The following are two specific examples.

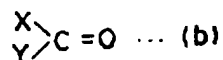
Method 1: A styryl compound can be synthesized using a method (Witting Reaction or Witting-Horner Reaction) in which a phosphonic acid ester expressed by Formula (a)

[0077] [Formula 20]



[0078] (where R is an alkyl group with 1 to 4 carbon atoms or a phenyl group, and R³⁷ through R⁴⁸ are the same as above) and a carbonyl compound expressed by Formula (b)

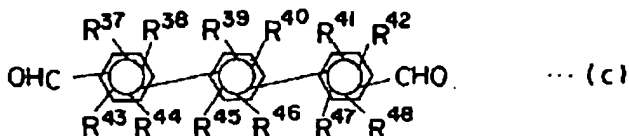
[0079] [Formula 21]



[0080] (where X and Y are the same as above) are condensed in the presence of a base.

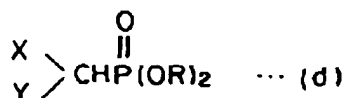
Method 2: A styryl compound can be synthesized using a method (Witting Reaction or Witting-Horner Reaction) in which a dialdehyde compound expressed by Formula (c)

[0081] [Formula 22]



[0082] (where R³⁷ through R⁴⁸ are the same as above) and a phosphonic acid ester expressed by Formula (d)

[0083] [Formula 23]



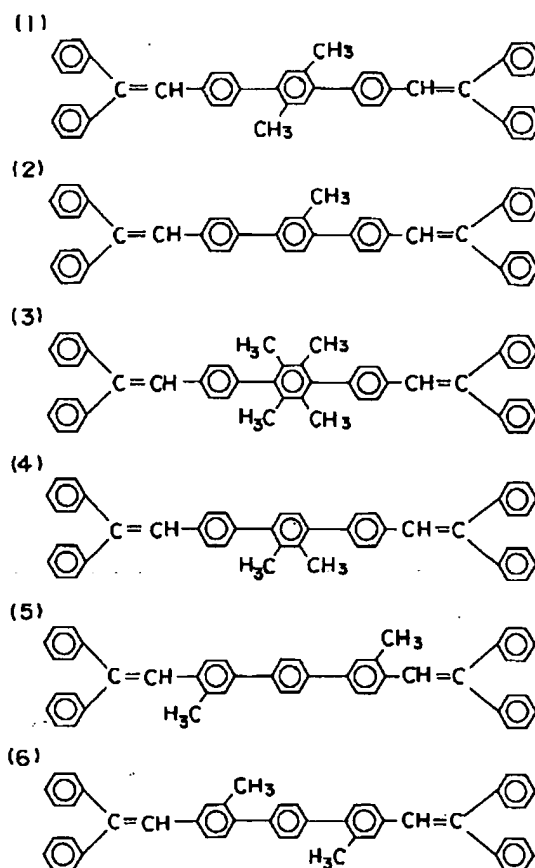
[0084] (where X and Y are the same as above) are condensed in the presence of a base.

[0085] Reaction solvents that can be used in the synthesis include hydrocarbons, alcohols and ethers. Specific examples include methanol, ethanol, isopropanol, butanol, 2-methoxyethanol, 1,2-dimethoxyethane, bis (2-methoxyethyl) ether, dioxane, tetrahydrofuran, toluene, xylene, dimethylsulfoxide, N,N-dimethylformamide, N-methylpyrrolidone, and 1,3-dimethyl-2-imidazolidinone. Preferred examples include tetrahydrofuran and dimethylsulfoxide. Preferred condensation agents include sodium hydroxide, potassium hydroxide,

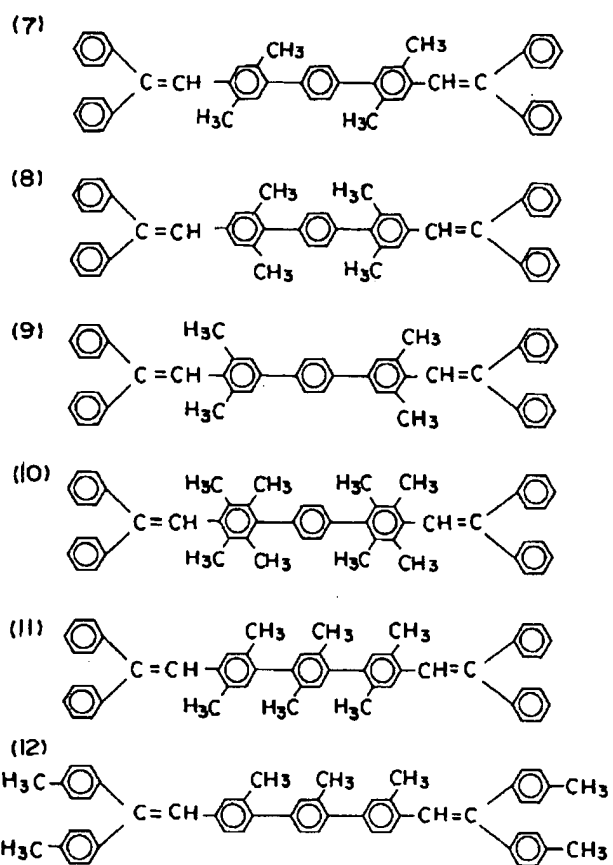
sodium amide, sodium hydride, n-butyl lithium, sodium methyllate, and potassium-t-butoxide. Especially preferred are n-butyl lithium and potassium-t-butoxide. The reaction temperature depends on the type of reaction materials being used is not rigorously defined. However, the reaction temperature usually ranges between 0°C and 100°C. A reaction temperature between 0°C and room temperature is especially preferred.

[0086] Specific examples of styryl compounds that can be used in the present invention include (1) through (26) below. The present invention, however, is not limited to these examples.

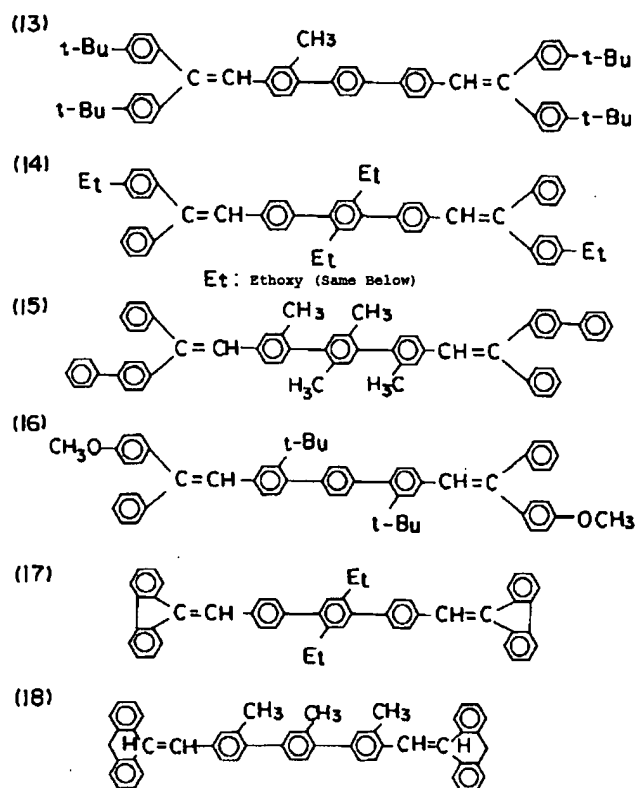
[0087] [Formula 24]



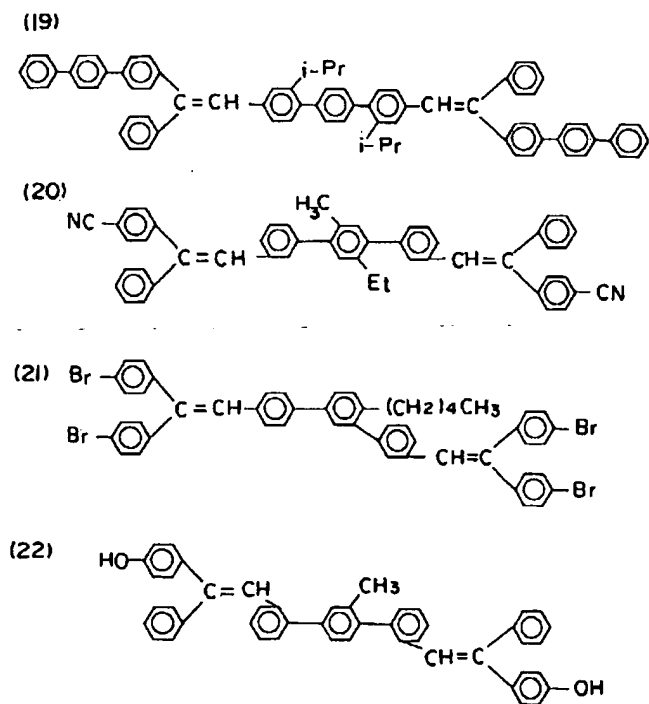
[0088] [Formula 25]



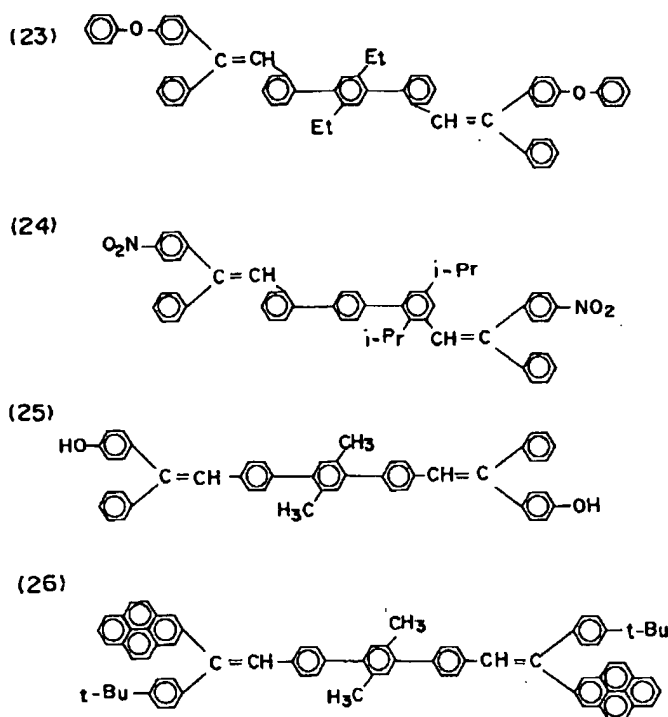
[0089] [Formula 26]



[0090] [Formula 27]

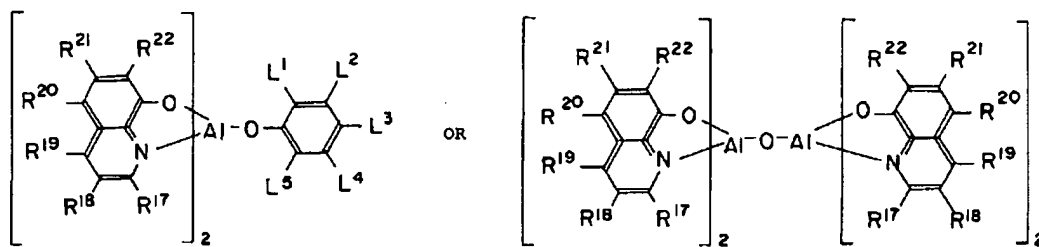


[0091] [Formula 28]



[0092] Aluminum complexes with the following structural formulas can also be used as a blue fluorescent material.

[0093] [Formula 29]

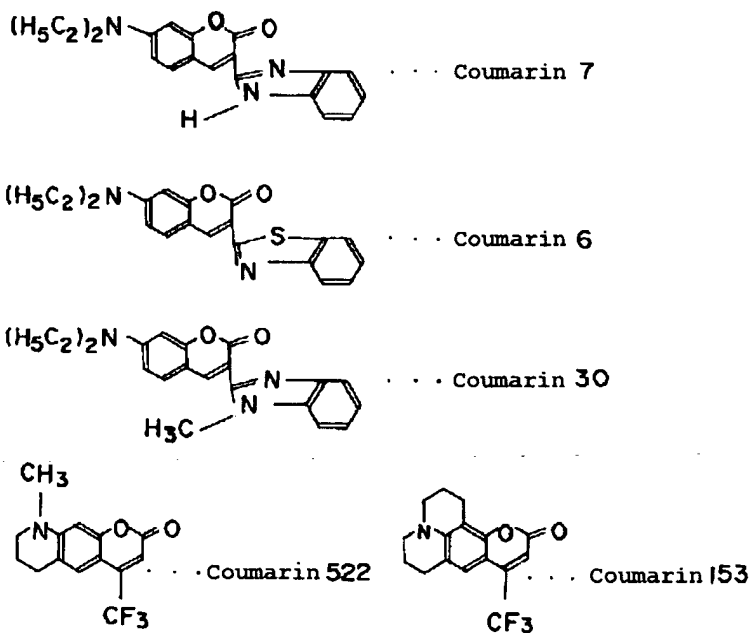


[0094] (In this formula, R^{17} through R^{19} separately denote a hydrogen atom or an alkyl group such as a methyl group, R^{20} through R^{22} separately denote a hydrogen atom, a halogen atom, an alpha-haloalkyl group, an alpha-haloalkoxy group, an amide group, a carbonyl group, a sulfonyl group, a carbonyloxy group, an oxycarbonyl group or an allyl group. L^1 through L^5 separately denote a hydrogen atom or a

hydrocarbon group with 1 to 12 carbon atoms. L^1 and L^2 as well as L^2 and L^3 can be bonded together to form an aromatic ring.) The thickness of the organic luminescent layers 31, 32, 33 range usually between 0.1 and 200 nm, preferably between 0.1 and 50 nm, and ideally between 1 and 40 nm.

[0095] There are no particular restrictions on the organic compounds used as the green fluorescent material in the present invention as long as the solid-state fluorescent peak wavelength is between 480 nm and 580 nm. An example is the coumarin derivative used as a laser pigment in European Unexamined Patent Application Publication No. 281,381. Specific examples include the following.

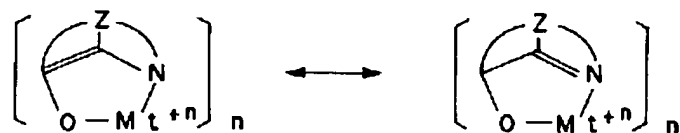
[0096] [Formula 30]



[0097] In addition, some of the organic compounds described in JP3-231970A and Japanese Patent Application No. 2-279304 satisfy the fluorescence conditions for a green fluorescent material. Preferred examples are metal complexes of 8-hydroxyquinone or a derivative

thereof. A specific example is a metal chelate oxynoid compound containing a metal chelate of oxine (8-quinolinol or 8-hydroxyquinoline). These high-performance compounds easily form a thin film. These oxynoid compounds satisfy the following structural formulas.

[0098] [Formula 31]



[0099] (In this formula, Mt denotes a metal, $n = 1-3$, Z denotes the atoms in different positions needed to complete at least two condensed aromatic rings.) Here, the metal expressed by Mt denotes a univalent, divalent or trivalent metal. Specific examples include alkali metals such as lithium, sodium or potassium, alkali earth metals such as magnesium or calcium, and earth metals such as boron or aluminum. Any univalent, divalent or trivalent metal commonly used in chelate compounds can be used here.

[0100] Z also denotes atoms in which at least two of the condensed aromatic rings form a heterocyclic ring consisting of an azole or a azine. A ring other than a condensed aromatic ring can be added if necessary. In order to avoid adding a bulky molecule whose performance cannot be improved, the number of atoms expressed by Z should be maintained at 18 or fewer.

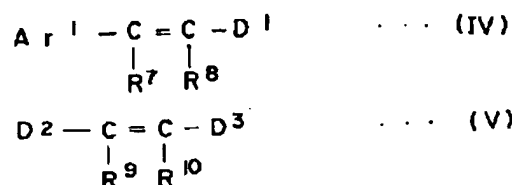
[0101] Specific examples of chelated oxynoid compounds include tris (8-quinolinol) aluminum, bis (8-quinolinol) magnesium, bis (8-quinolinol) zinc, bis (2-methyl-8-quinolinol) zinc, bis (benzo-8-

quinolinol) zinc, bis (2-methyl-8-quinolilate) aluminum oxide, tris (8-quinolinol) indium, tris (5-methyl-8-quinolinol) aluminum, 8-quinolinol lithium, tris (5-chloro-8-quinolinol) gallium, tris (2-methyl-8-quinolinol) gallium, bis (5-chloro-8-quinolinol) calcium, 5,7-dichloro-8-quinolinol aluminum, tris (5,7-dibromo-8-hydroxyquinolinol) aluminum, tris (7-propyl-8-quinolinol) aluminum, bis (8-quinolinol) beryllium, and bis (2-methyl-8-quinolinol) beryllium.

[0102] A white organic electroluminescent element used in the present invention contains a blue fluorescent material. There are no particular restrictions on the organic compounds used as the blue fluorescent material in the present invention as long as the solution-state fluorescent peak wavelength is between 380 nm and 480 nm. Preferred examples include at least one type selected from among the stilbene derivatives, distyrylarylene derivatives and tristyrylarylene derivatives described in the specification of Japanese Patent Application No. 5-129438. The stilbene derivatives are compounds with at least two aromatic rings, where the aromatic rings are bonded by a vinyl group or substituted vinyl group, and an electron-donating group for either the aromatic ring or the vinyl group. The distyryl arylene derivatives are compounds in which two aromatic rings are bonded to an arylene group via vinyl group or substituted vinyl group and in which the compound has an electron-donating group. The tristyryl arylene derivatives are compounds in which three aromatic rings are bonded to a trivalent aromatic ring via vinyl group or substituted vinyl group and in which the compound has an electron-donating group. In

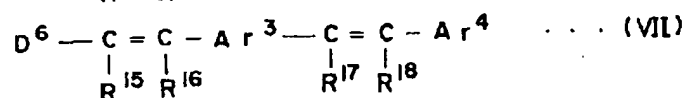
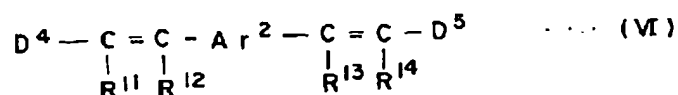
derivatives where the electron-donating group is in the molecular structure, the electron-donating group is preferably an alkoxy group with 1 to 10 carbon atoms, an aryloxy group with 6 to 10 carbon atoms or an amino group with a hydrocarbon group with 1 to 30 carbon atoms. These derivatives are preferably compounds expressed by Formula (IV) through Formula (X). Formula (IV) and Formula (V) express stilbene derivatives, Formula (VI) and Formula (VII) express distylyl arylene derivatives, and Formula (VIII) through Formula (X) denote tristylyl arylene derivatives.

[0103] [Formula 32]



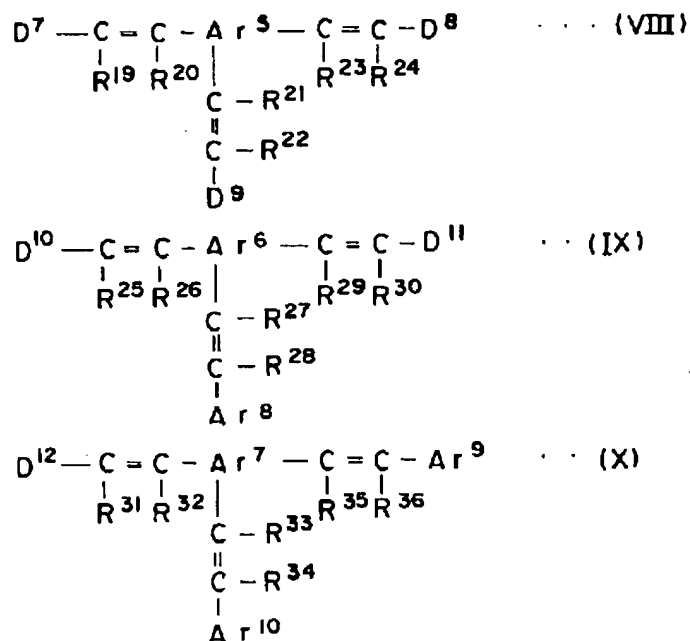
[0104] (In this formula, Ar¹ denotes an aryl group with 6 to 20 carbon atoms, R⁷ through R¹⁰ separately denote a hydrogen atom or an arylene groups with 6 to 20 carbon atoms. D¹ through D² separately denote an arylene group with 6 to 20 carbon atoms substituted by an electron-donating group or a condensed heterocyclic ring with 10 to 30 carbon atoms. Here, Ar¹ and R⁷ through R¹⁰ are either unsubstituted or substituted by an alkyl group with 1 to 10 carbon atoms, an alkoxy group with 1 to 10 carbon atoms, an aryloxy group with 6 to 10 carbon atoms, an aralkyl group with 6 to 10 carbon atoms, or an amino group with a hydrocarbon group containing 1 to 20 carbon atoms. These substituents can be bonded to each other to form a saturated or unsaturated five-member ring or six-member ring.)

[0105] [Formula 33]



[0106] (In these formulas, A^2 and A^3 separately denote arylene groups with 6 to 20 carbon atoms, and Ar^4 denotes an aryl group with 6 to 20 carbon atoms. R^{11} through R^{18} separately denote a hydrogen atom or an aryl group with 6 to 20 carbon atoms. Here, Ar^2 through Ar^4 as well as R^{11} through R^{18} are either unsubstituted or substituted by substituted by an alkyl group with 1 to 10 carbon atoms, an alkoxy group with 1 to 10 carbon atoms, an aryloxy group with 6 to 10 carbon atoms, an aralkyl group with 6 to 10 carbon atoms, or an amino group with a hydrocarbon group containing 1 to 20 carbon atoms. These substituents can be bonded to each other to form a saturated or unsaturated five-member ring or six-member ring. D^4 through D^6 separately denote an arylene group with 6 to 20 carbon atoms substituted by an electron-donating group or a condensed heterocyclic ring with 10 to 30 carbon atoms.)

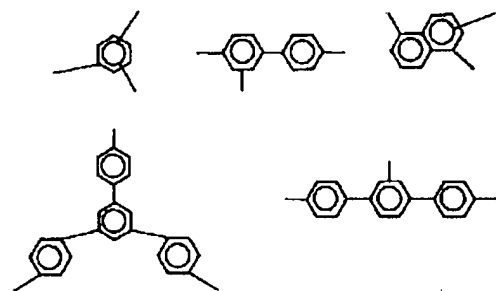
[0107] [Formula 34]



[0108] (In this formula, Ar⁵ through Ar⁷ separately denote a trivalent aromatic ring with 6 to 24 carbon atoms. Ar⁸ through Ar¹⁰ separately denote an aryl group with 6 to 20 carbon atoms. R¹⁹ through R³⁶ separately denote a hydrogen atom or an aryl group with 6 to 20 carbon atoms. D⁷ through D²⁰ separately denote an aryl group with 6 to 20 carbon atoms substituted by an electron-donating group or a condensed heterocyclic group with 10 to 30 carbon atoms. Here, Ar⁵ through Ar⁷ and R¹⁹ through R³⁶ are either unsubstituted or substituted by an alkyl group with 1 to 10 carbon atoms, an alkoxy group with 1 to 10 carbon atoms, an aryloxy group with 6 to 10 carbon atoms, an aralkyl group with 6 to 10 carbon atoms, or an amino group with a hydrocarbon group containing 1 to 20 carbon atoms. These substituents can be bonded to each other to form a saturated or unsaturated five-member or six-member ring.) Preferred examples of the aryl group in Formula (IV) through Formula (X) include a phenyl group, a biphenyloyl group, a

naphthyl group, a pyrenyl group, a terphenyloyl group, an anthranyl group, a tolyl group, a xylyl group, a stilbenyl group, a thienyl group, a bithienyl group, a thiophene group, a bithiophene group or a terthiophene group. Preferred examples of arylene groups include a phenylene group, a biphenylene group, a naphthylene group, an anthranylene group, a terphenylene group, a pyrenylene group, a stilbenylene group, a thienylene group or a bithienylene group. Examples of trivalent aromatic rings include the following.

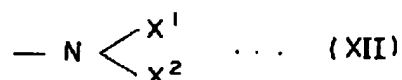
[0109] [Formula 35]



[0110] Among these substituents, the aryloxy group include a phenyloxy group, a biphenyloxy group, a naphthyloxy group, an anthranyloxy group, a terphenyloxy group, and a pyrenyloxy group. The alkyl groups include a methyl group, an ethyl group, an isoproyl group, a tertiary butyl group, a phenyl group or a benzyl group. The alkoxy groups include a methoxy group, an ethoxy group, an isopropoxy group, a tertiary butoxy group, and a pentyloxy group. Amino groups with a hydrocarbon group include a dimethyl amino group, a diethyl amino group, a diphenyl amino group, a phenylethyl amino group, a phenylmethyl amino group, a ditolyl amino group, an ethylphenyl amino

group, a phenylnaphthyl amino group, and a phenylbiphenyl amino group. D¹ through D¹² in Formula (IV) through Formula (X) are an aryl group with 1 to 20 carbon atoms substituted by an electron-donating group or a condensed heterocyclic group with 10 through 30 carbon atoms. The electron-donating group is preferably an alkoxy group with 1 to 10 carbon atoms, an aryloxy group with 6 to 20 carbon atoms or an amino group with a hydrocarbon group with 1 to 30 carbon atoms. A hydrocarbon group with 1 to 30 carbon atoms is especially preferred. A specific example of an amino group is shown in Formula (XII).

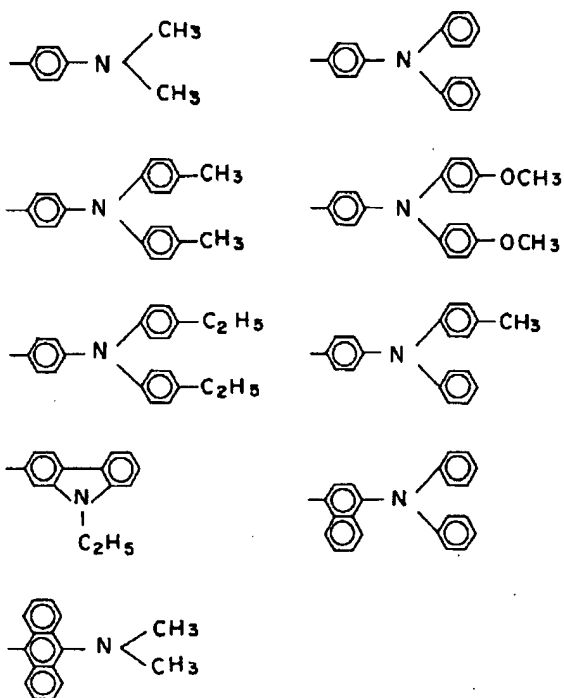
[0111] [Formula 36]



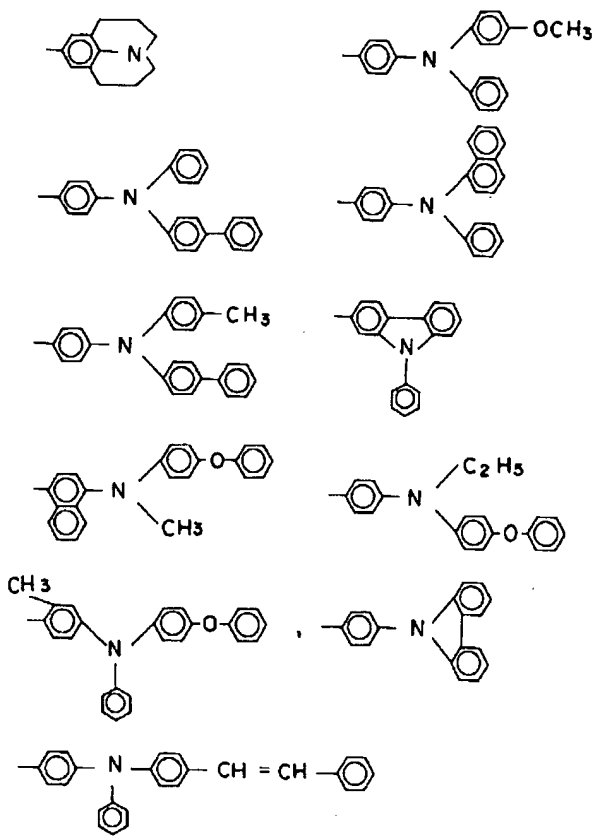
[0112] (In this formula, X¹ and X² separately denote an aryl group with 6 to 20 carbon atoms, an alkyl group with 1 to 10 carbon atoms, or an aralkyl group with 6 to 20 carbon atoms, where these groups can be bonded to each other to form a saturated or unsaturated cyclic structure. X¹ and X² can also be substituted by an alkyl group with 1 to 10 carbon atoms, an aralkyl group with 7 to 10 carbon atoms, an araloxy group with 6 to 10 carbon atoms, and an alkoxy group with 6 to 10 carbon atoms. If X¹ and X² are an aryl group substituted by an amino group expressed by Formula (XII), they can be bonded to each other to form an aromatic cyclic group containing nitrogen.) Examples of electron-donating groups include an aryloxy group such as a phenyloxy group, biphenyl group, naphthyloxy group, anthralyl group or terphenyl group; an alkoxy group such as a methoxy group, ethoxy group, isopropoxy group, tertiary butyloxy group or a pentyloxy group; or an

amino group with a hydrocarbon group such as a dimethyl amino group, diethyl amino group, diphenyl amino group, phenylmethyl amino group, phenylethyl amino group, phenylmethylethyl amino group, ditolyl amino group, ethylphenyl amino group, phenylnaphthyl amino group, and phenyl biphenyl amino group. The following are specific examples of D¹ through D¹².

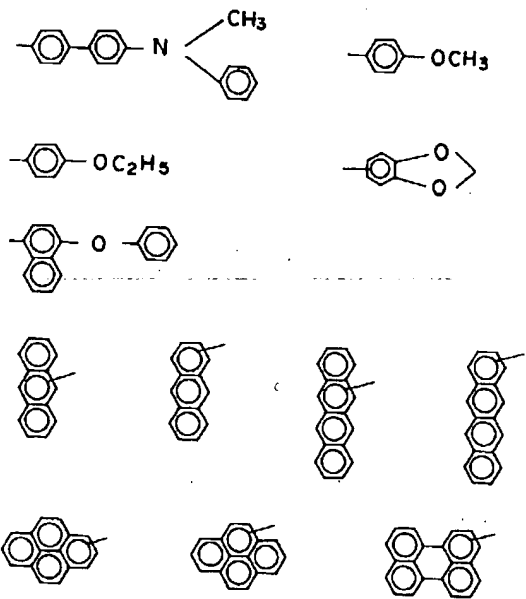
[0113] [Formula 37]



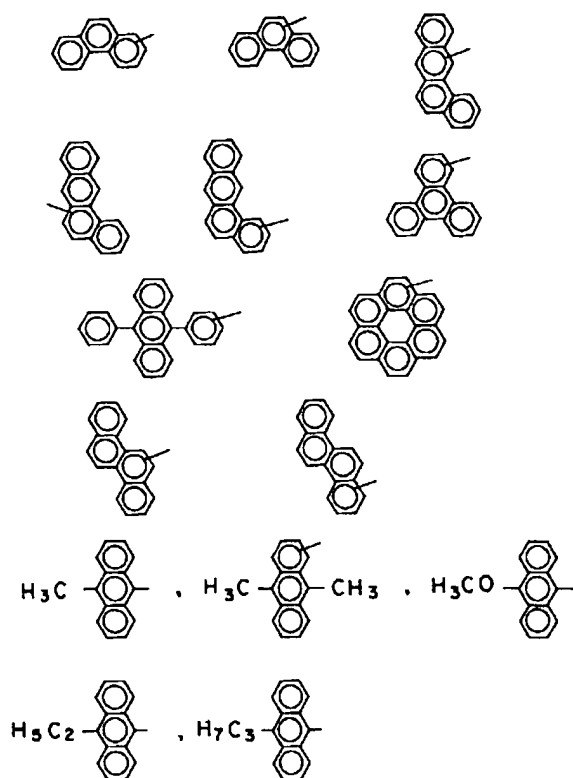
[0114] [Formula 38]



[0115] [Formula 39]

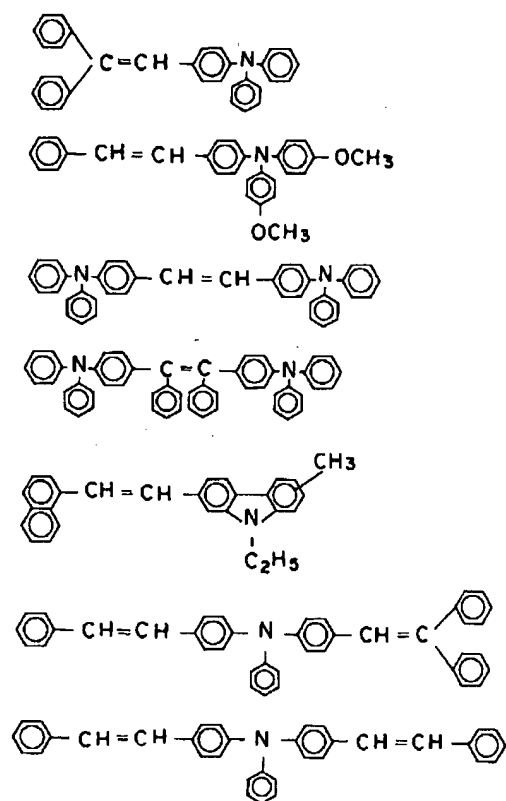


[0116] [Formula 40]

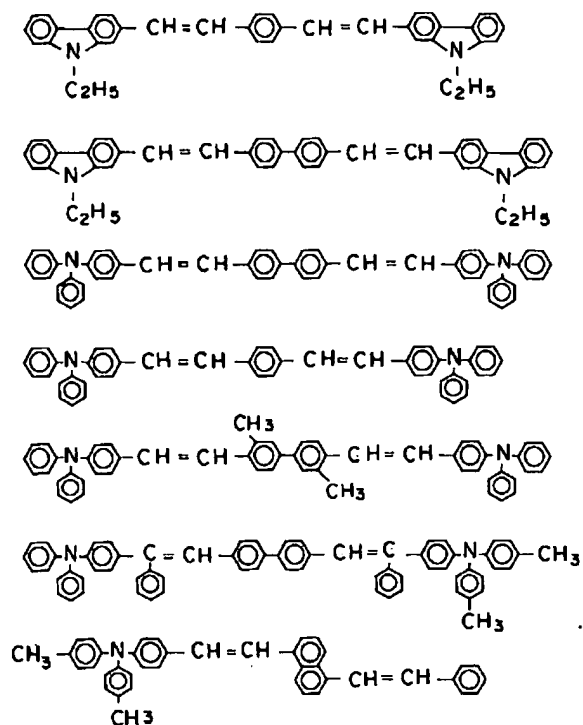


[0117] Specific examples of compounds expressed by Formulas (IV) through (X) include the following.

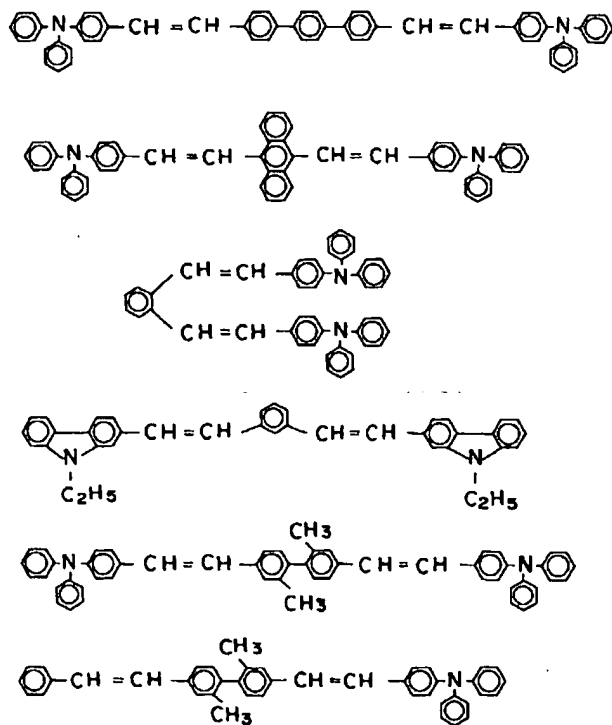
[0118] [Formula 41]



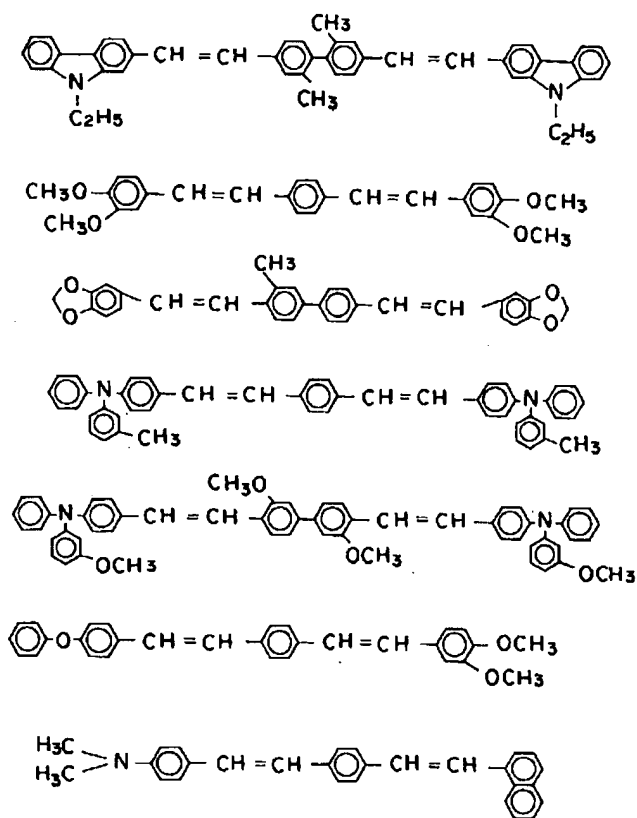
[0119] [Formula 42]



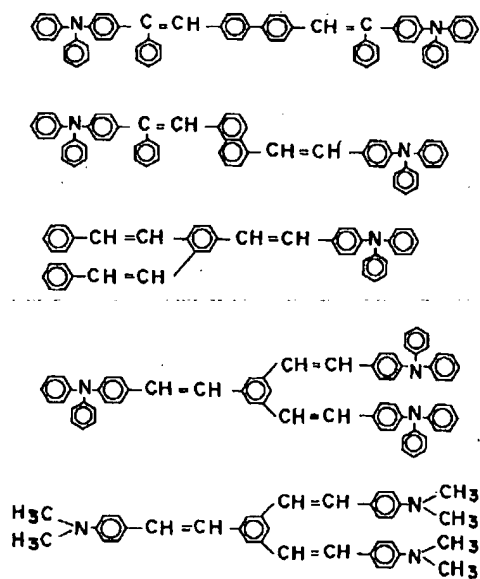
[0120] [Formula 43]



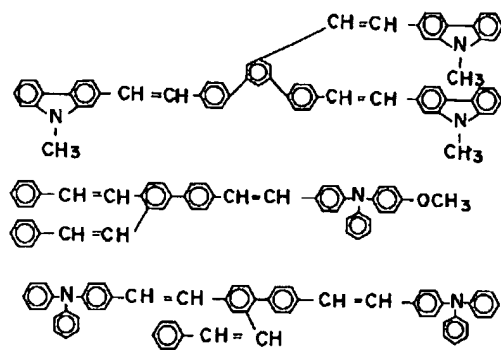
[0121] [Formula 44]



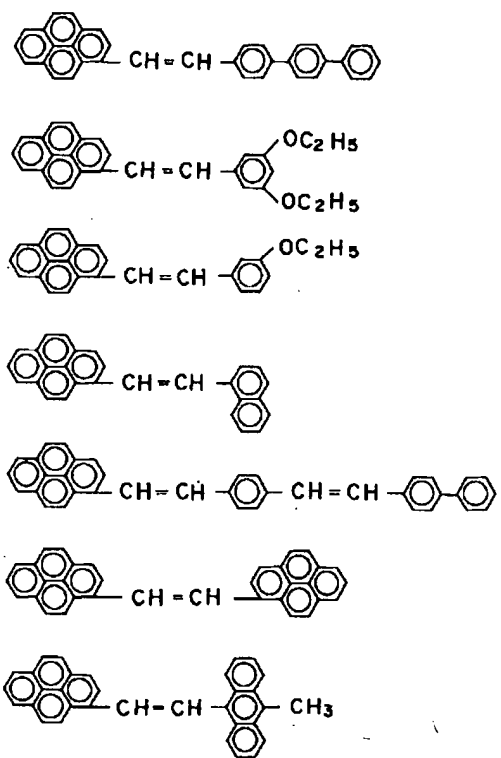
[0122] [Formula 45]



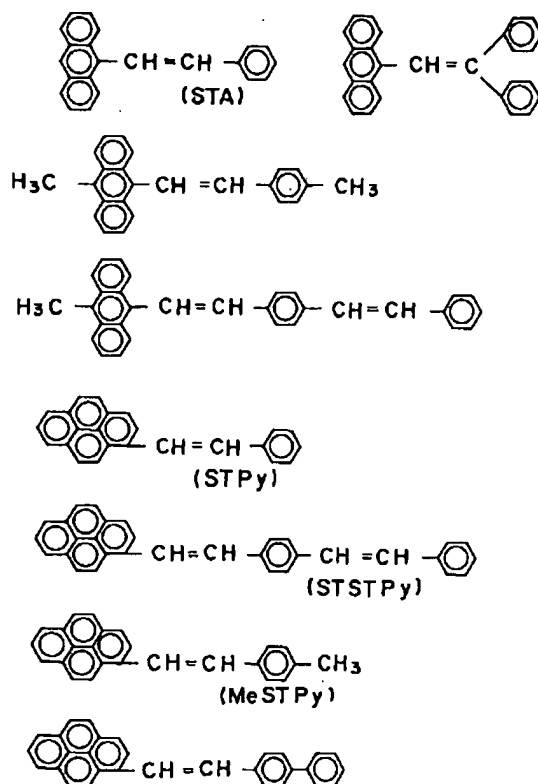
[0123] [Formula 46]



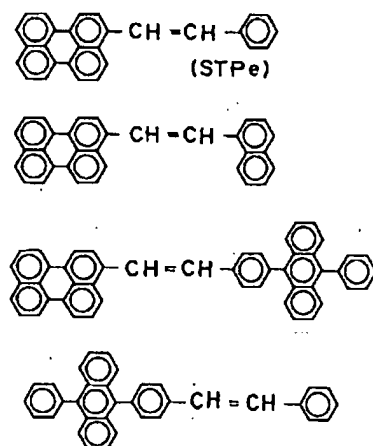
[0124] [Formula 47]



[0125] [Formula 48]



[0126] [Formula 49]

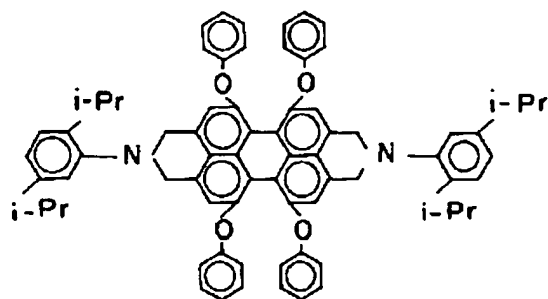


[0127] Other preferred blue fluorescent materials include polycyclic aromatics such as anthracene, perylene and coronene, and alkyl substituents thereof.

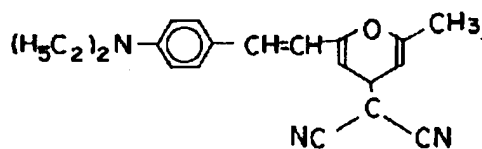
[0128] There are no particular restrictions on the organic compounds used as the green fluorescent material in the present invention as long as the solution-state fluorescent peak wavelength is between 480 nm and 580 nm. Green fluorescent materials for doping include coumarin compounds such as 3-(2'-benzimidazolyl)-7-N,N-diethylaminocoumarin (Coumarin 535), 3-(2-benzothiazolyl)-7-diethylaminocoumarin (Coumarin 540), 2,3,5,6-1H,4H-tetrahydro-8-trifluoromethylquinolidino-<9,9a,1-gh> coumarin (Coumarin 540A), 3-(5-chloro-2-benzothiazolyl)-7-diethylaminocoumarin (Coumarin 34), 4-trifluoromethyl-piperidino [3,2-g] coumarin (Coumarin 340), N-ethyl-4-trifluoromethyl-piperidino [3,2-g] coumarin (Coumarin 355), N-methyl-4-trifluoromethyl-piperidono [2,3-h] coumarin and 9-cyano-1,2,4,5-3H,6H,10H-tetrahydro-1-benzopyrano [9,9a1-gh] quinolidine-10-one (Coumarin 337); xanthine pigments such as 2,7-dichlorofluorecene; tetracene; and quinacridone compounds.

[0129] There are no particular restrictions on the organic compounds used as the red fluorescent material in the present invention as long as the solution-state fluorescent peak wavelength is between 580 nm and 650 nm. Examples include the dicyanomethylenepyrene derivatives, dicyanomethylenethiopyrene derivatives, fluorescein derivatives and perylene derivatives used as red laser pigments in European Unexamined Patent Application No. 0,281,381. Specific examples include the following.

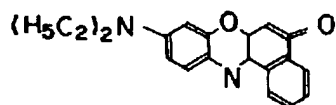
[0130] [Formula 50]



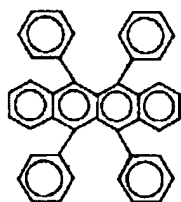
(Lumogen F Red)



(Dicyanomethylen Pyrane)



(Phenoxazone)



(Rubrene)

[0131] The organic compounds used to form the layers have to include 0.1 to 10 mol%, and preferably 0.5 to 5 mol%, of these organic compounds. 0.1 to 10 mol% is the concentration range in which concentration quenching does not occur.

(2) Hole-Injecting Layer

A hole-injecting layer is not a required element in the present invention, but use of a hole-injecting layer is preferred from the standpoint of improving light-emitting performance. A hole-injecting layer is a layer that assists in injecting holes into the fluorescent

layers. In this layer, the hole mobility is high and the ionized energy is usually 5.5 eV or less. The material used in the hole-injecting layer should be a material that is able to inject holes into the luminescent layers using a lower electric field. The hole mobility should be at least 10^{-6} cm²/V·sec at an electric field application of 10^4 to 10^6 V/cm. Any material that meets these performance requirements can be used as the hole-injecting material. These include light-conducting materials of the prior art, which are commonly used as charge transport materials for holes, as well as any material commonly used in the hole-injecting layers of electroluminescent elements.

[0132] Specific examples include triazole derivatives (USP 3,112,197), oxadiazole derivatives (USP 3,189,447), imidazole derivatives (JP 37-16096B), polyaral polyaralalkane derivatives (USP 3,615,402, USP 3,820,989, USP 3,542,544, JP 45-555B, JP 51-10983B, JP 51-93224A, JP 55-17105A, JP 56-4148A, JP 55-108667A, JP 55-156953A, JP 56-36656A), pyrazoline derivatives and pyrazolone derivatives (USP 3,180,729, USP 4,278,746, JP 55-88064A, JP 55-88065A, JP 49-105537A, JP 55-51086A, JP 56-80051A, JP 56-88141A, JP 57-45545A, JP 54-112637A, JP 55-74546A), phenylenediamine derivatives (USP 3,615,404, JP 51-10105B, JP 46-3712B, JP 47-25336B, JP 54-53435A, JP 54-110536A, JP 54-119925A), aralamine derivatives (USP 3,567,450, USP 3,180,703, USP 3,240,597, USP 3,658,520, USP 4,232,103, USP 4,175,961, USP 4,012,376, JP 49-35702B, JP 39-27577B, JP 55-114250A, JP 56-119132A, JP 56-22437A, West German Patent 1,110,518), amino-substituted chalcone derivatives (USP

3,526,501), oxazole derivatives (USP 3,257,203), stylylantracene derivatives (JP 56-46234A), fluorenone derivatives (JP 54-110837A), hydrazone derivatives (USP 3,717,462, JP54-59143A, JP 55-52063A, JP 55-52064A, JP 55-46760A, JP 55-85495A, JP 57-11350A, JP 57-148749A, JP 2-311591A), stilbene derivatives (JP 61-210363A, JP 61-228451A, JP 61-14642A, JP 61-72255A, JP 62-47646A, JP 62-36674A, JP 62-10652A, JP 62-30255A, JP 60-93445A, JP 60-94462A, JP 60-174749A, JP 60-175052A), silazane derivatives (US 4,950,950), polysilanes (JP 2-204996A), aniline copolymers (JP 2-282263A), and the conductive macromolecular oligomers (especially the thiophene oligomers) disclosed in JP 1-211399A. Any one of these can be used as the hole-injecting layer material. However, the following are preferred: porphyrin compounds (JP 63-295696A), and aromatic tertiary amine compounds (USP 4,127,412, JP 53-27022A, JP 54-58445A, JP 54-149634A, JP 54-64299A, JP 55-79450A, JP 55-144250A, JP 56-119132A, JP 61-295558A, JP 61-98353A, JP 63-295695). The aromatic tertiary amine compounds are especially preferred. Specific examples of porphyrin compounds include porphyrin, 1,10,15,20-tetraphenyl-21H,23H-porphin copper (II), 1,10,15,20-tetraphenyl-21H,23H-porphin zinc (II), 5,10,15,20-tetrakis(pentafluorophenyl)-21H,23H-porphin, silicon phthalocyanine oxide, aluminum phthalocyanine chloride, phthalocyanine (no metal), dilithium phthalocyanine, copper tetramethyl phthalocyanine, copper phthalocyanine, chromium phthalocyanine, zinc phthalocyanine, lead phthalocyanine, titanium phthalocyanine oxide, Mg phthalocyanine, and copper octamethyl phthalocyanine. Specific examples of aromatic

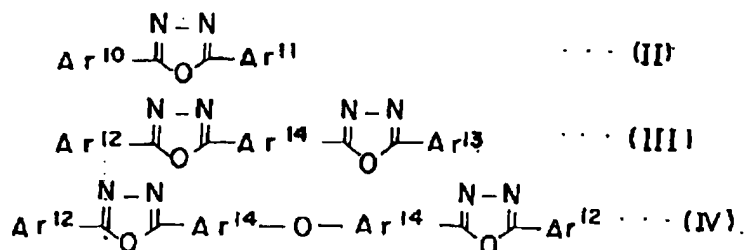
tertiary amine compounds and stylyl amine compounds include N,N,N',N'-tetraphenyl-4,4'-diaminophenyl, N,N'-diphenyl-N,N'-bis-(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (TPD), 2,2-bis(4-di-p-tolylaminophenyl) propane, 1,1-bis (4-di-p-tolylaminophenyl) cyclohexane, N,N,N',N'-tetra-p-tolyl-4,4'-diaminophenyl, 1,1-bis (4-di-p-tolylaminophenyl)-4-phenylcyclohexane, bis (4-dimethylamino-2-methylphenyl) phenylmethane, bis (4-di-p-tolylaminophenyl) phenylmethane, N,N'-diphenyl-N,N'-di(4-methoxyphenyl)-4,4'-diaminophenyl, N,N,N',N'-tetraphenyl-4,4'-diaminophenylether, 4,4'-bis (diphenylamino) quadriphenyl, N,N,N-tri (p-tolyl) amine, 4-(di-p-triamino)-4'-[4(di-p-triamino)stylyl] stilbene, 4-N,N-diphenylamino-(2-diphenylvinyl) benzene, 3-methoxy-4'-N,N-diphenylamino stilbenzene; N-phenylcarbazole; 4,4'-bis [N-(1-naphthyl)-N-phenylamino] biphenyl (NPD) with two condensed aromatic rings in the molecule as described in USP 5,061,569, or 4,4',4"-tris [N-(3-methylphenyl)-N-phenylamino] triphenylamine (MTDATA) in which a triphenyl amine unit is connected to three starbursts as described in JP4-308688A. In addition to these aromatic dimethylidene compounds, inorganic compounds such as p-Si and p-SiC can be used as the hole-injecting layer material. The hole-injecting layer can be formed by creating a thin film with these compounds using any method common in the art such as the vacuum deposition method, the spin coat method, the cast method or the LB method. There are no restrictions on the thickness of the hole-injecting layer, but the thickness is usually 5 nm to 5 μ m. The hole-injecting layer can consist of layers of one or more materials. Hole-

injecting layers made of different compounds can also be laminated. The organic semiconductor layer assists in injecting holes or electrons into the luminescent layers. It should have a conductivity greater than 10^{-10} S/cm. The material used in the organic semiconductor layer can be a conductive oligomer containing a thiophene oligomer or an aral amine oligomer, or a conductive dendrimer containing an aral amine dendrimer.

(3) Electron-Injecting Layer

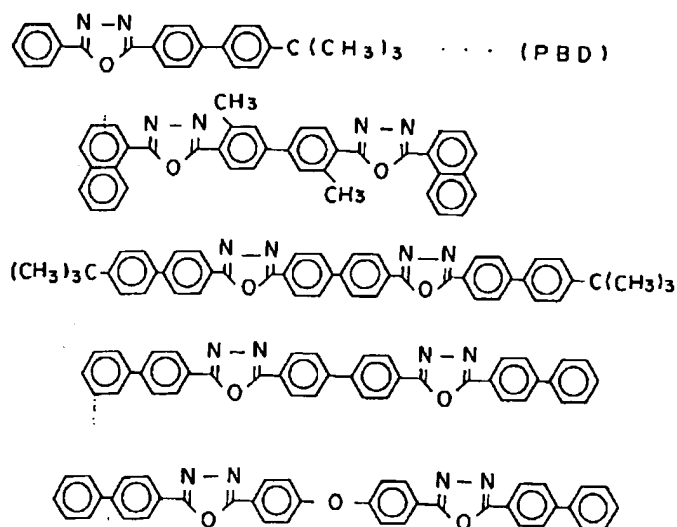
An electron-injecting layer helps to inject electrons into the luminescent layers. It therefore has a high degree of electron mobility. The electron-injecting layer also serves an adhesion improving layer and is made of a material that adheres well, especially to the negative electrode. The material used in the electron-injecting layer is preferably a metal complex of 8-hydroxyquinone or a derivative thereof, or a oxadiazole derivative. A metal complex of 8-hydroxyquinone or a derivative thereof is ideal as the material used in the adhesion improving layer. Specific examples of a metal complex of 8-hydroxyquinone or a derivative thereof include a metal chelate oxynoid compound containing a chelate of oxine (especially 8-quinolinol or 8-hydroxyquinoline). Examples of oxadioazole derivatives include the electron transferring compounds expressed by Formula (II), Formula (III) and Formula (IV).

[0133] [Formula 51]



[0134] (In these formulas, Ar¹⁰ through Ar¹³ denote substituted or unsubstituted aryl groups, Ar¹⁰ and Ar¹¹ as well as Ar¹² and Ar¹³ are both the same or different, and Ar¹⁴ denotes a substituted or unsubstituted arylene group.) Here, the aryl groups include a phenyl group, biphenyl group, anthralyl group, phenanthryl group, pyrenyl group or perilenyl group; and the arylene groups include a phenylene group, naphthalene group, biphenylene group, anthracenylene group, perilenylene group or pyrenylene group. The substituent can be an alkyl group with 1 to 10 carbon atoms, an alkoxy group with 1 to 10 carbon atoms, and a cyano group. The electron transferring compound should have good thin film forming properties. Specific examples of the electron transferring compounds include the following.

[0135] [Formula 52]



[0136] [Working Examples]

The following is a more detailed explanation of the present invention with reference to working examples.

[Working Example 1]

An ITO electrode was formed on top of a 25 mm x 75 mm x 1.1 mm glass substrate to a thickness of 100 nm to form a transparent support substrate. This was rinsed ultrasonically for five minutes in isopropyl alcohol, rinsed for another five minutes in pure water, and then rinsed for yet another five minutes in isopropyl alcohol. The transparent support substrate was attached to the substrate holder in a commercially available vacuum deposition device (Nippon Vacuum Technologies), seven molybdenum resistance heat boards were prepared, and into the vacuum chamber reduced to 1×10^{-4} Pa were added 500 mg of 4,4',4''-tris [N-(m-tolyl)-N-phenylamino]-triphenylamine (MTDATA) expressed by Formula 53, 200 mg of N,N'-diphenyl-N,N'-bis-(3-methylphenyl)-[1,1'-biphenyl]-4,4'-diamine (TPD), 200 mg of 4,4'-bis

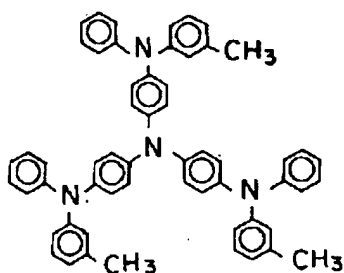
(2,2-diphenylvinyl)biphenyl (DVPBi), 200 mg of 4,4'-bis [2-{4-(N,N-diphenylamino)phenyl}vinyl] biphenyl (DPAVBi), 200 mg of quinacridone (QN) expressed by Formula 53, 200 mg of DCM expressed by Formula 53 and, finally, 100 mg of tris (8-hydroxyquinoline) aluminum (Alq). First, the board with the MTDATA was heated and the MTDATA laminated on the substrate to form a hole-injecting layer with a thickness of 60 nm. Next, the board with the TPD was heated to vaporize the TPD and deposit an electron-transferring layer with a thickness of 20 nm. Next, the board with the DPVBi and the board with the DPAVBi were simultaneously deposited on top of the electron-transferring layer to form a 20 nm-thick blue luminescent layer (mixture weight ratio of DPVBi to DPAVBi = 40 : 1). Next, the board with the DPVBi and the board with the QN were simultaneously heated and vaporized to deposit a green luminescent layer with a thickness of 10 nm (mixture weight ratio of DPVBi to QN = 40: 1). Next, the board with the DPVBi and the board with the DCM were simultaneously heated and vaporized to deposit a red luminescent layer with a thickness of 10 nm (mixture weight ratio of DPVBi to DCM = 40 : 3). Finally, Alq (electron-injecting layer) was deposited to 20 nm. This was removed from the vacuum tank, a stainless steel mask was placed over the luminescent layers, and this was attached once again to the substrate holder. 0.5 g of Ag wire was placed in a tungsten basket, and 1 g of a Mg ribbon was placed on another molybdenum board. The pressure inside the vacuum tank was reduced to 1×10^{-4} Pa, the Mg was deposited at a rate of 1.8 nm/s and the Ag was deposited at the same time at a rate of 0.1 nm/s to form a

negative electrode. When 8 V of voltage was applied to the ITO positive electrode and the Mg:Ag negative electrode, uniform white light was generated by the element [CIE chromacity coordinates (0.36, 0.34)]. The element had a current density at 8 V of 2.1 mA/cm², and a brightness of 200 cd/m². The efficiency was 3.74 (lm/W).

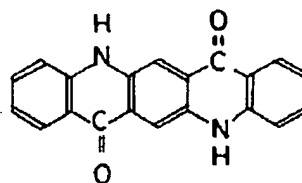
[0137] [Working Example 2]

An element was manufactured in exactly the same way as the first working example except that the green luminescent layer is a mixed luminescent layer consisting of Alq and the coumarin in Formula 53 (where the weight ratio of Alq to coumarin is 40 : 1). The element had a current density of 1.9 mA/cm² at 8 V, a brightness of 196 cd/m², and generated uniform white light (CIE chromacity coordinates: 0.34, 0.33). The efficiency was 3.96 (lm/W).

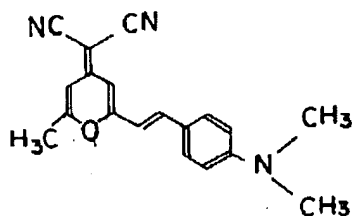
[0138] [Formula 53]



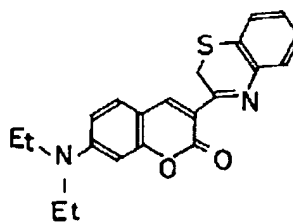
MTDATA



QN



DCM

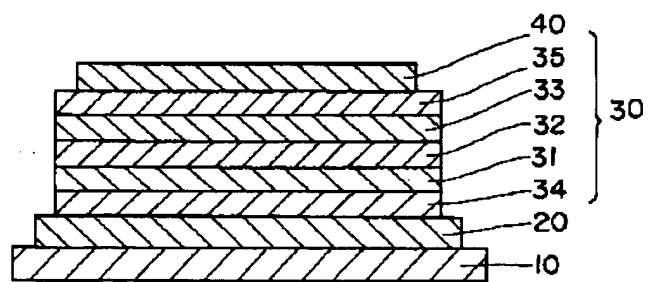


Coumarin

[0139] [Working Example 3]

An element was manufactured in the same manner as the first working example except that the operation in the first working example is repeated twice in the manufacture of luminescent layers with a thickness of 5 nm. In other words, the configuration of this element is the following: glass substrate / ITO / MTDATA / TPD / blue luminescent layer / green luminescent layer / red luminescent layer / blue luminescent layer / green luminescent layer / red luminescent layer / electron-injecting layer / Mg : Ag. The element had a current density of 3.7 mA/cm² at 8 V, a brightness of 250 cd/m², and generated uniform white light (CIE chromacity coordinates: 0.36, 0.34). The efficiency was 2.56 (lm/W).

[FIG 1]



[0140] [Effect of the Invention]

As explained above, the present invention provides a highly efficient organic electroluminescent device that able to generate very bright white light.

[Brief Explanation of the Figure]

[FIG 1] A simplified cross-sectional view of the organic electroluminescent device in a working example of the present invention.

[Key to the Figure]

- 10: Substrate
- 20: Substrate Electrode
- 30: Organic Layer Containing Luminescent Layers
- 31: Blue Luminescent Layer
- 32: Green Luminescent Layer
- 33: Red Luminescent Layer
- 34: Hole-Injecting Layer
- 35: Electron-Injecting Layer
- 40: Opposing Electrode